

A Stochastic Bulk Model for Turbulent Collision and Coalescence of Cloud Droplets

by

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B.A. University of Michigan, 1994

M.A. Wayne State University, 2010

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Abstract

We propose a mathematical procedure to derive a stochastic parameterization for the bulk warm cloud micro-physical properties of collision and coalescence. Unlike previous bulk parameterizations, the stochastic parameterization does not assume any particular droplet size distribution, all parameters have physical meanings which are recoverable from data, all equations are independently derived making conservation of mass intrinsic, the auto conversion parameter is finely controllable, and the resultant parameterization has the flexibility to utilize a variety of collision kernels. This new approach to modelling the kinetic collection equation (KCE) decouples the choice of a droplet size distribution and a collision kernel from a cloud microphysical parameterization employed by the governing climate model. In essence, a climate model utilizing this new parameterization of cloud microphysics could have different distributions and different kernels in different climate model cells, yet employ a single parameterization scheme.

This stochastic bulk model is validated theoretically and empirically against an existing bulk model that contains a simple enough (toy) collision kernel such that the KCE can be solved analytically. Theoretically, the stochastic model reproduces all the terms of each equation in the existing model and precisely reproduces the power law dependence for all of the evolving cloud properties. Empirically, values of stochastic parameters can be chosen graphically which will precisely reproduce the coefficients of the existing model, save for some ad-hoc non-dimensional time functions. Furthermore values of stochastic parameters are chosen graphically. The values selected for the stochastic parameters effect the conversion rate of mass cloud to rain. This conversion rate is compared against (i) an existing bulk model, and (ii) a detailed solution that is used as a benchmark.

The utility of the stochastic bulk model is extended to include hydrodynamic and turbulent collision kernels for both clean and polluted clouds. The validation and extension compares the time required to convert 50% of cloud mass to rain mass, compares the mean rain radius at that time, and used detailed simulations as benchmarks. Stochastic parameters can be chosen graphically to replicate the 50% conversion time in all cases. The curves showing the evolution of mass conversion that are generated by the stochastic model with realistic kernels do not match corresponding benchmark curves at all times during the evolution for constant parameter values. The degree to which the benchmark curves represent ground truth, i.e. at-

mospheric observations, is unknown. Finally, among alternate methods of acquiring parameter values, getting a set of sequential values for a single parameter has a stronger physical foundation than getting one value per parameter, and a stochastic simulation is preferable to a higher order detailed method due to the presence of bias in the latter.

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1 | Introduction

Clouds are among the most poorly understood components of climate models. Physical processes in clouds have length scales which span nine orders of magnitude (Pruppacher & Klett 1997). Computers are not capable of performing the calculations necessary to resolve all of the microphysical processes that occur in a cloud that is fully contained in a model's grid cell (Devenish & Bartello 2012). Therefore, approximations and simplifications to the dynamic equations that represent these microphysical processes are necessary when these processes provide climate models with macroscopic cloud information such as the onset of precipitation and radar reflectivity (Franklin et. al. 2007).

Climate models are a set of partial differential equations that are numerically solved for prognostic variables. A set of Navier-Stokes equations on a rotating sphere and conservation equations describes the atmosphere. Prognostic variables describing the atmosphere include air velocity, temperature, surface pressure, and water vapour (Gill 1982).

Climate models use horizontal and vertical grids to partition the atmosphere. A general circulation model may have horizontal grids as fine as ($O(10^5\text{m})$). Cloud resolving models (CRM) are typically used more for research than for weather or climate predictions. The horizontal distance between the grid boundaries ($O(10^2\text{-}10^3\text{m})$) in a CRM is less than the length scales of clouds ($O(10^3\text{-}10^4\text{m})$). Therefore, the grid cells in a CRM are either cloudy or cloud free and CRM explicitly resolves macroscale convective flows (Blossey et. al. 2007).

Clouds are a major source of uncertainty in predictions of climate. Because CRMs resolve processes that atmospheric general circulation models

(AGCM) do not resolve, CRMs can address some of the uncertainty. Output parameters of CRMs can be used as a reference for parameterizations of processes that are smaller than the grid sizes of atmospheric general circulation models. Microphysical processes in clouds are not resolved by CRMs. These unresolved processes introduce uncertainties and challenges to our understanding of the effects that cloud microphysics has on climate predictions. CRMs can also be coupled with AGCM to model some processes that are not resolved by AGCM (Blossey et al. 2007). This is the technique of super-parameterization of cloud resolving cumulus parameterizations (Randall et al. 2003). One type of uncertainty is the choice of the size distribution of water droplets in clouds. The droplet size distributions are not necessarily the same for differing cloud types (Pruppacher & Klett 1997) and consequently for different cells in a CRM.

A sequence of three general stages result in the production of rainfall at the Earth’s surface: condensation and nucleation, collision and coalescence, and precipitation (Grabowski & Wang 2013). Each of these stages is modelled as a suite of physical processes. The processes of collision and coalescence are dominant after condensation and nucleation when water droplets are several microns in radii, and before the droplets are large enough to have a terminal velocity which exceeds typical cloud updrafts (Franklin 2008). Typically CRMs use bulk rate equations which are a set of coupled differential equations for rain and cloud aggregates that simplify the microphysical processes.

Reliable determinations of the onset of rainfall and the radar reflectivity in clouds are dependent on increasing the accuracy of models that evolve the cloud’s droplet size distribution (DSD) (Pruppacher & Klett 1997). The simplest models evolve an initial DSD using only collision and coalescence terms: self-collection (cloud and rain), auto conversion, and accretion. Auto conversion is the only one of these processes that appears in each of the differential equations in the coupled set. It is also the one most strongly influenced by turbulence which makes it the most difficult to accurately model (Shaw 2003). Over the past 40+ years, more research has focused on parameterizing auto-conversion, particularly the auto conversion producing rain mixing ratio, than the other processes. When a full suite of collision and coalescence parameterizations were derived, it is a common practice to make the remaining auto conversion processes to be functions of the parameterized rain auto conversion (Khairoutdinov & Kogan 2000, Seifert & Beheng 2001, Franklin 2008), thereby enforcing conservation of mass.

Current bulk models are dependent on an assumed DSD (Seifert & Beheng 2001, Liu & Daum 2004, Liu, Daum, McGraw & Wood 2006), are tied to a specific collision kernel (Franklin 2008), or use a single parameterization to represent a variety of cloud types and environs (Khairoutdinov & Kogan 2000). The assumptions regarding a DSD, a collision kernel, a cloud’s type age or environ give closure to their model’s underlying equations. Milbrandt and Yau used a gamma distribution for number concentration, but their model included ice particles as well (Milbrandt & Yau 2005*a*). Their mixed phase model treated the ad-hoc gamma shape parameter prognostically and resulted in a three moment scheme (Milbrandt & Yau 2005*b*).

The use of stochastic methods in cloud microphysics is fairly new besides a couple of articles known to the author. Posselt et al. (2010) and Van Lier-Walqui et al. (2012) represented the uncertainty in radar reflectivity via a random process. Krueger (1993) (Krueger 1993) used a power law distribution to model the variability of entrainment plume size (Posselt & Vukicevic 2010, Lier-Walqui, M. & Posselt 2012). This was further developed by Krueger et al. (1997) to include multiple events and simulate a cumulus cloud (Krueger, Chwen-Wei & McMurtry 1997), and then developed by Su et al (1998) to include Kolmogorov inertial range scalings (Kolmogorov 1991) and droplet growth (Su et. al, 1998) . These stochastic models use probability distributions to model uncertainty (‘sample-based’ stochastic models). The stochastic bulk parameterization developed herein uses stochastic differential equations (SDE) and assumes no specific probability distributions. This method can be identified as an ‘SDE-based’ stochastic model. The author is unaware of any other ‘SDE-based’ stochastic model used to represent microphysical processes in clouds. The advantage of an ‘SDE-based’ stochastic model over a ‘sample-based’ stochastic model is that the latter requires multiple simulations to calculate statistics on the results while the former contains the variability intrinsically and requires only one simulation for a given set of parameters.

We propose a new methodology to derive bulk cloud microphysics equations from the kinetic collection equation in the case of the collision and coalescence of liquid water in warm clouds, but in principle it can be applied to more general situations. The partitioning of ice particles into categories, such as size, shape or density, can lead to different conversion rates (Morrison & Milbrandt 2015). Perhaps the uncertainty of ice particle distributions could be reshaped into the uncertainty of stochastic processes, as we present here for collision and coalescence. Our strategy is a new two-fold approach to modelling the kinetic collection equation (KCE) utilizing two assumptions

in a novel way. The uncertainties associated with the droplet size spectrum are reshaped into uncertainties of a stochastic process. We accomplish this by partitioning the droplet spectrum into two large bins representing cloud and rain aggregates, and representing droplet densities as the sum of a state space mean and a set of random fluctuations from the mean. Simultaneously, we use a Taylor approximation for the collision kernel around the centres of masses of bulk cloud and rain aggregates which allows the resulting parameterization to be independent of the collision kernel, i.e. the parameterization is developed for an arbitrary collision kernel. We close the high order moments by way of Ornstein-Uhlenbeck like processes with fixed means and fixed standard deviations. Two-moment equations are thus obtained for cloud and rain mass and number aggregates with stochastic parameters associated with three collision processes: cloud self-collection, auto conversion, and accretion.

Our method possesses several desirable features. We derive a stochastic bulk model of collision and coalescence that does not assume any particular probability distribution, gives physical meanings to all model parameters, closes the higher moments of the model's underlying equations in a novel way using stochastic processes, promotes conservation of mass through the independent derivation of each equation in the coupled set of ODEs, and provides for an unprecedented controlling of the auto conversion parameter. The absence of any assumed probability distribution is particularly important because the distributions in different cells in a climate model may be a function of the cloud type (maritime vs. continental or clean vs. polluted) and age and are not necessarily the same in each climate model cell.

The values of these physically meaningful parameters are constrained by conservation of mass and consistency of number, and the auto conversion parameter is further constrained by the threshold value between cloud and rain droplets. Appropriate parameter values can be acquired graphically within a defined 'region of validity' as a subset of the stochastic parameter space. By freeing the bulk model of collision and coalescence from a single droplet size distribution and a single collision kernel, different stochastic parameters can be chosen for different cloud types (maritime vs. continental or clean vs. polluted) and cloud ages.

The stochastic bulk model is validated against detailed simulations of the KCE and compared to the bulk model by Seifert and Beheng (2001) which uses a prescribed droplet distribution and a piece-wise polynomial collision kernel. The stochastic parameterization precisely reproduces the power law

characteristics of Seifert and Beheng’s model when their collision kernel is used. Their model can be derived using a particular subset (SB subset) of stochastic parameter values within the ‘region of validity.’ These two bulk parameterizations are benchmarked against results from detailed simulations using Bott’s (1998) Linear Flux Method (LFM). Two metrics are used to benchmark these bulk models: the time at which 50% of cloud mixing ratio is converted to rain, and the mean rain radius at that time. When the stochastic model produces results that closely match results from the LFM, the stochastic parameters are contained within the intersection of the ‘SB subset’ and the ‘region of validity’ for a clean, maritime cloud, but not for a polluted cloud. Sensitivity tests indicate that the stochastic model can be used with a time step as large as 30 seconds without significantly compromising accuracy.

The utility of the stochastic model is extended to use realistic collision kernels. Simulations based on a hydrodynamic kernel and two turbulent kernels are run for each of the polluted and clean cloud types. For each of these simulations we also acquire stochastic parameter values graphically and successfully match the 50% percent mass conversion of cloud mass to rain mass with the conversion time of the LFM. An alternate method of acquiring parameter values is proposed: getting a separate parameter value when each collision process is dominant. Additionally, two other methods, besides the graphical method, of acquiring parameter values are compared using partial results. One uses stochastic simulations of droplet collisions and the other uses a higher order detailed method.

The dissertation is organized as follows. In Chapter 2 we survey the literature in the areas of collision kernels and their various types, the KCE and its differing interpretations, the stochastic simulation of collision and coalescence, and the detailed and bulk evolution of the droplet spectrum. In Chapter 3 we derive the stochastic bulk model of collision and coalescence. In Chapter 4 the stochastic bulk model is validated theoretically and empirically by comparison with an existing bulk model which was derived using an analytic kernel, and the results from the evolution of the DSD using that model. In Chapter 5 the utility of the model is extended to include realistic kernels: a hydrodynamic and two turbulent kernels, and parameter values are obtained graphically. In Chapter 6 values for two of the stochastic parameters (cloud self-collection and auto conversion) are obtained by a detailed bin-pair spectral method. In Chapter 7 the results of the stochastic bulk model are summarized and preliminary results are given which offer direction on acquiring parameter values using a method other than the graphical one.

In Chapter 8 the new bulk model is summarized in the context of existing models and relative to alternate parameter acquisition methods, the impact of the research in the broader scientific community is given, and the next steps in the development of future work is noted.

2 | Literature Survey

2.1 The Kinetic Collection Equation (KCE)

The kinetic collection equation (KCE), known by many names such as the stochastic collection equation, the coagulation equation, Smoluchowski's equation, the scalar transport equation, or the master equation, was introduced by Smoluchowski in 1916 (Smoluchowski 1916) & (Pruppacher & Klett 1997):

$$\frac{\partial n(x, t)}{\partial t} = \frac{1}{2} \int_0^x n(x - x', t) n(x', t) K(x - x', x') dx' - \int_0^\infty n(x, t) n(x', t) K(x, x') dx' \quad (2.1)$$

where x and t are the droplet mass and time, respectively: $n(x, t)$ is the number concentration, a density function: and $K(x, x')$ is the collision-coalescence kernel, a rate function, so that $n(x, t)n(x', t)K(x, x')$ is the rate-density of collision-coalescence between two droplets of mass x and x' , respectively. When the KCE evolves a probability distribution, $f(x, t)dt$ is the probability that a given droplet has mass x during the interval $t + dt$. Here we are representing the evolution of number concentration which is the first moment ($\int x f(x, t) dx$) of a probability distribution function $f(x, t)$. The first integral on the r.h.s. is the gain integral and gives the increase of $n(x, t)$. The second integral is the loss integral and gives the decrease of $n(x, t)$ (Bott 1998, Wang 2007).

The KCE driven by a constant kernel, and with the probability distribu-

tion function, and an arbitrary initial distribution, subject to physically real conditions, was shown to have a unique and continuous solution for all time (Melzak 1957). The KCE with a product kernel and an arbitrary initial distribution, was shown to have uniqueness and existence for finite time (McLeod 1964). When the sum of masses kernel is used in the KCE, a solution was shown to exist, be unique, and have limiting characteristics (as $t \rightarrow \infty$) for an exponential initial condition (Golovin 1963). In 1968 Scott derived solutions for the KCE driven by each of these kernels and with arbitrary initial conditions (Scott 1968). Drake and Wright in 1972 derived solutions for the KCE containing some linear combinations of these three kernels (Drake & Wright 1972). That same year Drake derived solutions for moments of the KCE with certain linear combinations of the analytic kernels (Drake 1972). In 1974 Long fit collection rates over droplet radii to get coefficients for polynomial kernels in the spirit of linear combinations of these three collection kernels.

The stochastic interpretation of the KCE was introduced in 1955 by considering the random fluctuations in time which lead to some droplets growing faster than other droplets and producing a broader spectrum (Telford 1955). Telford performed simulations using a ‘double delta’ initial distribution (90% of droplets at $10\mu\text{m}$ radius and 10% at $20\mu\text{m}$). Using the KCE to evolve the droplet size distribution is referred to as the ‘stochastic growth’ method vs. a ‘continuous growth’ method that evolves all ‘collector’ droplets uniformly. The ‘stochastic growth’ theory produced a broader droplet size spectrum (with a few ‘high growth’ drops) than what was produced when with the ‘continuous growth’ theory was applied, i.e. where each ‘collector’ droplet uniformly grows as a result of coalescence with one of the smaller sized droplets (Telford 1955). E. Berry showed the ‘stochastic growth’ theory produced a broader spectrum than the ‘continuous growth’ theory when a gamma initial distribution was applied (Berry 1967). Chin and Neiburger (1972) simulated the evolution of the droplet size spectrum using a Monte Carlo method and a bi-disperse initial spectrum. They noted that there is nothing inherently stochastic about the kinetic collection equation (i.e. the rate function and the distribution function are deterministic), but they show that the mean drop mass from the ensemble of Monte Carlo simulations is compatible with the drop size distribution evolved by the ‘stochastic growth’ model (Chin & Neiburger 1972).

Under the assumptions that droplet densities are uncorrelated and same size droplets do not collide, the KCE was derived from first principles, and the probability $P(n, m; t)$ of finding n droplets of size m at time t approaches a

Poisson distribution for each m -size droplet (Gillespie 1972). Gillespie then rigorously defined three alternative interpretations of the KCE when it is used to model the collision and coalescence process. When discussing expressions related to these interpretations, $0 \leq y < \infty$. In his interpretations referred to as ‘continuous,’ $\sum_{y=0}^{\infty} n(x)K(x,y)dt$ is defined as the number of droplets of mass y that a droplet of mass x will collect in time dt . The ‘quasi-stochastic’ interpretation defines $\sum_{y=0}^{\infty} n(x)K(x,y)dt$ as the fraction of droplets of mass x that will collect a droplet of mass y in time dt . The ‘fully stochastic’ interpretation defines $\sum_{y=0}^{\infty} n(x)K(x,y)dt$ as the probability that any droplets of mass x will collect a droplet of mass y in time dt (Gillespie 1975*b*). In 1974 Bayewitz and collaborators claimed that for a sufficiently populated simulation run by a constant collision kernel; the correlations diminish, the variance and covariances become small, the KCE becomes deterministic and the KCE is highly accurate in representing collision and coalescence (Bayewitz et. al., 1974).

2.2 Collision Kernels

A collision kernel $K(x, x')$ is a non-negative, symmetric rate function where x and x' are masses, and is the rate of collision and coalescence of droplets of size x with droplets of size x' . Given a droplet number concentration $n(x, t)$, the “pure stochastic model” describes $n(x, t)K(x, x')dt$ as the probability that a droplet of size x will ‘collect,’ or collide and coalescence, with any other size droplet in time dt (Gillespie 1975*b*). There are three types of collision kernels: analytic, hydrodynamic, and turbulent. When an analytic or ‘toy’ kernel is used, the Equation 2.1 can be solved analytically (Drake & Wright 1972, Long 1974). The other two types add enough complexity that this integro-differential equation (KCE) can only be solved numerically.

2.2.1 Analytic Kernel

There are a few forms of an analytic collision kernel. It can take a constant form $K_A(x, x') = A$, or be proportional to the sum or product of the masses

of colliding droplets: $K_B(x, x') = B(x + x')$ and $K_C(x, x') = Cxx'$, respectively, where A, B, C are non-negative constants. Golovin (1963) introduced the sum of masses kernel. Long (1974) used the sum of masses kernel for radii larger than $50 \mu\text{m}$. The multiplicative kernel was shown to have a singularity in finite time (McLeod 1964). The constant kernel does not represent physically real collision processes, but its simplicity encourages its use (Melzak 1957, Bayewitz, Terushalmi, Katz & Shinnar 1974). The analytic kernel can also be composed of linear combinations of these forms (Drake & Wright 1972, Long 1974):

$$K_{ABC}(x, x') = A + B(x + x') + Cxx' \quad (2.2)$$

Three families of kernels have been proposed: (i) $B=aA$ and $C=0$, (ii) $C=2aB$, $A=0$, and (iii) $A=1$, $B=a$, and $C=a^2$ (Drake & Wright 1972, Drake 1972, Long 1974).

The sum of masses kernel is considered to be an acceptable approximation for droplets with a radius greater than $500\mu\text{m}$. However, it has been used when the error in Stokes flow becomes unacceptable: $30\mu\text{m}$ (Golovin 1963), $40\mu\text{m}$ (Seifert & Beheng 2001), $50\mu\text{m}$ (Long 1974). The sum of masses kernel has been used to validate detailed, bin-pair interaction methods (Bott 1998, Wang, Xue & Grabowski 2007) and bulk methods (Tzivion, Feingold & Levin 1987, Seifert & Beheng 2001).

Seifert and Beheng (2001) used a piece-wise combination of the sum of masses kernel by Golovin (1963) and quadratic polynomial kernel by Long (1974) for their bulk parameterizations:

$$K(x, y) = \begin{cases} k_c(x^2 + y^2), & x \text{ and } y < x^* \\ k_r(x + y), & x \text{ or } y \geq x^* \end{cases} \quad (2.3)$$

where $k_c = 9.44 \times 10^9 \text{ cm}^3 \text{g}^{-2} \text{s}^{-1}$, $k_r = 5.78 \times 10^3 \text{ cm}^3 \text{g}^{-1} \text{s}^{-1}$, and $x^* = 40 \mu\text{m}$ (the threshold between cloud droplets and rain droplets). This kernel was also used in a subsequent paper by Seifert and Stevens (2010).

2.2.2 Hydrodynamic Kernel

Hydrodynamic kernels are more accurate models of realistic collision rates because they consider gravitation. They take the form of the product of a cylindrical volume and an efficiency factor where the height of the cylinder

is the difference in terminal velocities of the two droplets:

$$K_H(r_1, r_2) = \pi R^2 |v_{t1} - v_{t2}| E_{\text{coll}} \quad (2.4)$$

The terminal velocities of the droplets are v_{t1} and v_{t2} , and $R = r_1 + r_2$ ($r_1 > r_2$) is the sum of the radii of the two colliding droplets (Meyer & Deglon 2011). The collection efficiency E_{coll} is the product of the collision efficiency E_{clsn} and the coalescence efficiency E_{coal} . Collision efficiency is the ratio of the effective droplet sweep out volume to the actual droplet sweep out volume. Coalescence efficiency is the ratio of the number of coalescences to the number of collisions in a unit volume for a unit time interval, and is taken to be unity due to inconclusive experimental results (Pruppacher & Klett 1997).

The hydrodynamic kernels commonly use a collision efficiency by Hall (1980) and terminal velocities by Beard (1976) such as in the works by Seeβelberg et. al. (1996), Bott (1998), and Wang et. al. (2007). Hall combined collision efficiency data from three regimes. When the larger droplet r_1 has a radius less than or equal to $30\mu\text{m}$, then the collision efficiency follows from the application of Stokes flow with a slip flow correction as shown by Davis (1972) and Jonas (1972). For $r_1 \leq 300\mu\text{m}$ and a radius ratio of 0.60 or less, the collision efficiencies follow from a linear superposition of droplet flow fields. Shafir and Gal-Chen (1971) used this superposition principle when $50\mu\text{m} \leq r_1 \leq 300\mu\text{m}$, and Lin and Lee (1975) used it when $20\mu\text{m} \leq r_1 \leq 200\mu\text{m}$. For droplets with a radius $10\mu\text{m}$ - $70\mu\text{m}$, Klett and Davis (1973) used a modified Oseen flow. Beard used Klett and Davis's data for $40 \leq r_1 \leq 70\mu\text{m}$ and a radius ratio greater than 0.60 and assigned a collision efficiency of unity for $r_1 > 70\mu\text{m}$. Seeβelberg et. al. (1996) modified Hall's collision efficiency table by allowing collision efficiencies greater than unity for collector droplets with a radius $70 \leq r_1 \leq 300\mu\text{m}$ due to wake capture effects (Lin & Lee 1975).

A hydrodynamic kernel with collision efficiencies by Hall (1980) modified by Seeβelberg et. al. (1996) and terminal velocities by Beard (1976) will be used in Chapters 5 and 6 to expand the stochastic bulk model and acquire stochastic parameters, respectively. This kernel will be used in both chapters for the hydrodynamic case, and also as a base for the multiplicative turbulent enhancements for collisions in which the collector droplet has a radius within the domain of the turbulent enhancements. The tabulated collision efficiencies are presented in Table A.1, and a contour plot of the hydrodynamic kernel containing these efficiencies is given in Figure 2.1.

Figure 2.1: Contour Plot: Hydrodynamic Collision Rates

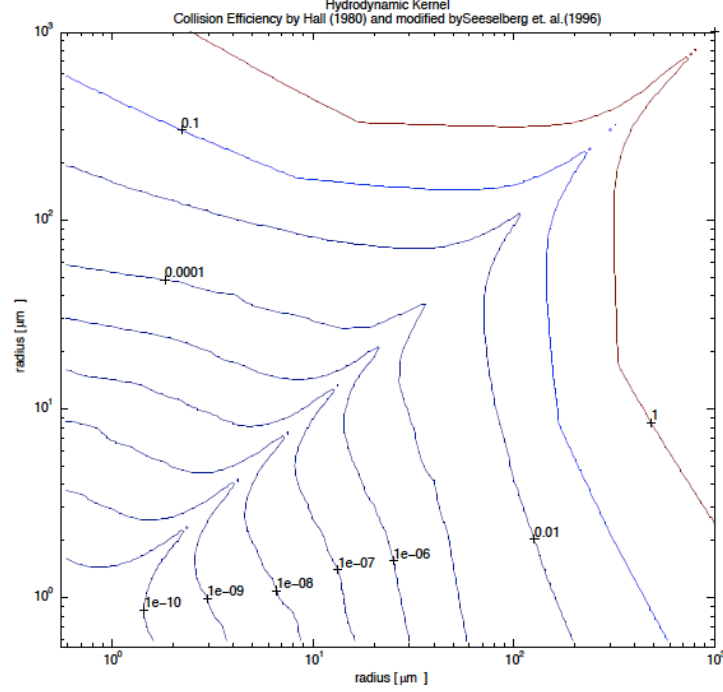


Figure 2.1: Shown is a contour plot of collision rates using a hydrodynamic kernel with Hall's table of collision efficiencies that were modified by Seeßelberg et. al. (1996) to include wake effects suggested by Lin and Lee (1975) and terminal velocities by Beard (1976).

2.2.3 Turbulent Kernel

In a turbulent flow the velocity will vary randomly from one location to another, at a given location it will vary chaotically over time, and at a single point and given instant it will vary randomly from one realization to the next realization. Turbulence occurs when the velocities at 'small' scales are not determined by the macroscopic flow (Batchelor 1953). Turbulence is also characterized by length and velocity scales that are large compared to the kinematic viscosity of the fluid. Turbulence is graphically described by coherent, rotating structures called 'eddies' which exist at many different length scales (Pope 2000).

Eddies of the largest length scales absorb energy from the mean flow. This

energy cascades through eddies of decreasing size solely through inertial forces and is dissipated into internal energy by eddies at the smallest scales through the process of molecular diffusion. As such, kinetic energy is dissipated into internal energy by shear stress (Wyngaard 2010).

Smoluchowski introduced a model for turbulent shear in 1917 (Pruppacher & Klett 1997). Saffman and Turner added fluid acceleration to Smoluchowski's shear term and included a term for gravity (Saffman & Turner 1956). Wang and collaborators included a coupling term to the turbulent kernel and re-derived the other terms using a spherical model instead of the cylindrical model (Wang, Wexler & Zhou 1998). The gravitational term is the same in both conceptual models. The spherical interpretation was shown to be the correct formulation for the turbulent phenomena and gives the following turbulent collision kernel:

$$K_w(r_1, r_2) = 2\sqrt{2\pi}R^2 \left[\underbrace{\frac{1}{15}R^2 \frac{\bar{\epsilon}}{\nu}}_{\text{shear}} + \underbrace{\left(1 - \frac{\rho}{\rho_0}\right)^2 (\tau_1 - \tau_2)^2 \overline{\left(\frac{Du}{Dt}\right)^2}}_{\text{fluid acceleration}} + \right. \\ \left. \underbrace{2\left(1 - \frac{\rho}{\rho_0}\right)^2 \tau_1^2 \tau_2^2 \overline{\left(\frac{Du}{Dt}\right)^2} \frac{R^2}{\lambda_D^2}}_{\text{coupling}} + \underbrace{\frac{\pi}{8}\left(1 - \frac{\rho}{\rho_0}\right)^2 (\tau_1 - \tau_2)^2 g^2}_{\text{gravity}} \right]^{1/2} \quad (2.5)$$

where $\bar{\epsilon}$ is the mean turbulent dissipation rate, ν is the kinematic viscosity, ρ and ρ_0 are the densities of the fluid and droplets, τ_1 and τ_2 are the relaxation times of the colliding droplets, $\overline{\left(\frac{Du}{Dt}\right)^2}$ is the x component of the fluid acceleration, λ_D is the longitudinal Taylor microscale of fluid acceleration, and g is the acceleration due to gravity (Wang et al. 1998).

Shear, fluid acceleration, and gravity all share a conceptual similarity in that they are all dependent on the relative velocities of the pre-collision droplets. Considering the relative velocity phenomena as a single term, the general form of a turbulent collision kernel is

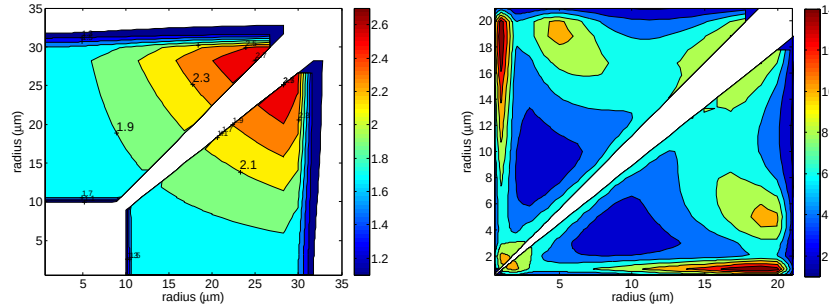
$$K_T(r_1, r_2) = 2\pi R^2 \overline{|w_r|} g(R), \quad (2.6)$$

where $w_r = \frac{\mathbf{w} \cdot \mathbf{R}}{|\mathbf{R}|}$ is the radial component of the relative velocity between two droplets, is a function of the terminal velocities, and the overbar indicates the mean. The droplet's relative velocity and separation vector are given by $\mathbf{w}$ and $\mathbf{R}$, respectively. The radial distribution function (coupling term) is

given by $g(R)$ Franklin (2007). The coupling function given by Franklin (2007) is a ratio of the probability of a pair of source droplets colliding in a given simulation to the probability of the source droplets colliding when they are uniformly distributed spatially, the specific form of which is not relevant to this research and is therefore omitted.

Collision efficiencies in a turbulent collision kernel need to be calculated with a set a conditions and assumptions different from the gravitational case. These efficiencies cannot simply be substituted in place of gravitational efficiencies because the turbulent effect on the geometric volume would be ignored (Pruppacher & Klett 1997). Franklin (2007) parameterized coupling and radial relative velocity using direct numerical simulation. Wang and collaborators tabulated a turbulent enhancement factor from velocity gradient tensors and Lagrangian accelerations in a laboratory experiment.

**Figure 2.2: Contour Plot:
Turbulent Enhancement of Collision Rates**



(a) Parameterized Turbulent Collision Rates (b) Tabulated Turbulent Collision Rates

Figure 2.2: The contour plot (a) shows the parameterized turbulent enhancement by Franklin et. al. (2007) at a turbulent energy dissipation rate of $1500 \text{ cm}^2\text{s}^{-3}$ (Taylor-based Reynolds number: 50.5) applied to collector droplets with a radii 10-30 μm . The contour plot (b) shows the tabulated enhancement factor by Pinsky et. al. (2008) at a turbulent energy dissipation rate of $1000 \text{ cm}^2\text{s}^{-3}$ and a Taylor-based Reynolds number of 20000, and was applied to collector droplets with radii between 1-21 μm . Both turbulent models enhance a hydrodynamic kernel with collision efficiencies by Hall (1980) and as modified by Seeβelberg et. al. (1996).

Parameterized Kernel

Franklin et. al. (2007) parameterized radial relative velocity and clustering by fitting an exponential curve to data acquired from direct numerical simulation (DNS). Turbulent radial relative velocity is normalized by the product of the gravitational relative velocity ($w_{r,grav}$) and the Reynolds number (R_λ) of the turbulence:

$$\frac{\overline{|w_r|}}{w_{r,grav}R_\lambda} = a_v e^{b_v \frac{v_{t1}v_{t2}}{v_\nu^2}} + c_v. \quad (2.7)$$

The fitting constants are $a_v = 0.0195$, $b_v = 0.0145$, and $c_v = 0.0084$. The terminal velocities are v_{t1} and v_{t2} , and v_ν is the Kolmogorov characteristic velocity. Clustering is normalized by the Reynolds number:

$$\frac{g(R)}{R_\lambda} = a_c e^{b_c \frac{v_{t1}v_{t2}}{v_\nu^2}} + c_c. \quad (2.8)$$

The fitting constants for the clustering parameterization are $a_c = -0.0139$, $b_c = -0.3331$, and $c_c = 0.0394$.

Franklin et. al. (2007) compared a hydrodynamic kernel with the turbulent enhancement given by the parameterizations in Equations (2.7 and 2.8). The comparison was done on a 2D droplet radius space at turbulent dissipation rates of 100, 500, 1000, 1500 cm^2s^{-3} . The turbulent enhancement is given by $\Psi(r_1, r_2)$:

$$\Psi(r_1, r_2) = \frac{\overline{|w_r|}g(R)}{w_{r,grav}} = R_\lambda^2 (a_v e^{b_v \frac{v_{t1}v_{t2}}{v_\nu^2}} + c_v) (a_c e^{b_c \frac{v_{t1}v_{t2}}{v_\nu^2}} + c_c).$$

The ratio of the turbulent enhanced geometric kernel to a hydrodynamic geometric kernel is:

$$T(r_1, r_2) = \frac{\pi R^2 w_{r,grav} \Psi(r_1, r_2) E_{clsn} E_{coal}}{\pi R^2 w_{r,grav} E_{clsn} E_{coal}} \quad (2.9)$$

where E_{clsn} are the collision efficiencies described by Seeβelberg et. al. (1996) and E_{coal} is the coalescence efficiency set to unity. The enhancement factor $\Psi(r_1, r_2)$ includes the ratio of turbulent to gravitational collision efficiencies since the factors comprising it were fitted to DNS that considered that difference. Values for the parameterized enhancement factor over the domain (10-30 μm) are presented in Figure 2.2a.

The factor of two that is present in the turbulent geometric kernel Equation 2.6 but not in the hydrodynamic geometric kernel Equation 2.4 does not appear in their ratio of the turbulent to hydrodynamic geometric parameterizations. Although they do not confirm or acknowledge this factor of two, the omission may be due to it being intrinsically contained in the DNS data and thus being implicitly present in the parameterization $\Psi(r_1, r_2)$.

Tabulated Kernel

Through a series of five articles Pinsky, Khain and groups of collaborators developed a method for calculating collision efficiencies and collision kernel enhancement factors in turbulent flows. The efficiencies and enhancement factors depended on values of the turbulent dissipation rates and Reynolds numbers that would be expected in atmospheric clouds.

Time series of velocity gradient tensors and Lagrangian accelerations at high Reynolds numbers were obtained from laboratory experiments and theoretical studies. Moments of these time series were collected at points centred in an array of unit volumes each containing uniform flow properties. The moments of velocity shears and flow accelerations were used to parameterize turbulent effects so that values of shear ($S_{ij}(i, j = 1, 2, 3)$ is a tensor of turbulent velocity gradients) and fluid acceleration (A_i) at droplet centres could be used to model droplet-droplet interactions, with x_i and x_j as positions of pre-collision droplet pairs, in equations of motion.

Continuing the notation used by Pinsky, Khain, et. al. (2008), a droplet's velocity affected by the motion of the other droplet (hydrodynamic droplet interaction - HDI), an unaffected droplet velocity, and a droplet's terminal velocity are given by $\check{V}_i$, $\hat{V}_i'$, and V_t , respectively. The equations of motion for a droplet's position, the non-disturbed velocity (noHDI), and disturbed velocity (HDI) are

$$\begin{aligned} \frac{dx_i}{dt} &= S_{ij} + V_t \delta_{3i} + \hat{V}_i' + \check{V}_i \\ \hat{V}_i' \left(\frac{1}{\tau} \delta_{ij} + S_{ij} \right) &= A_i + V_t S_{3i} \\ \frac{d\check{V}_i}{dt} &= -\check{V}_i \left(\frac{1}{\tau} \delta_{ij} + S_{ij} \right) + \frac{1}{\tau} u_j^*(x_i), \text{ respectively,} \end{aligned} \quad (2.10)$$

where τ is the characteristic relaxation time, δ_{ij} is a delta-function, and $u_j^*(x_i)$ is the fluid velocity induced by the droplet located at x_j and disturbing

the droplet located at x_i . When $\check{V}_i = 0$ the droplets do not affect the velocity fields of the surrounding fluid. In the absence of shear and fluid acceleration, $\check{V}_i = \hat{V}_i' = 0$ and droplet motion reverts back to the gravitational case.

Collision efficiency is the effect that HDI has on collisions of droplets that are simultaneously in the same collision sphere. This is represented as the ratio of the area through which the collected droplet passes and collides with the collector droplet to $A_g = \pi(r_1 + r_2)^2$ where r_1 and r_2 are the radii of the droplets. When HDI pushes the collected droplet away from the collector droplet, we have $E_{\text{coll}} < 1$. When hydrodynamic phenomena such as wake effect pull the collected droplet towards the collector droplet, we have $E_{\text{coll}} > 1$.

Pinsky, Khain, and collaborators (2007, 2008) calculated this ratio by integrating backwards, without HDI ($\check{V}_i=0$), from all points of possible droplet-droplet collisions to generate a manifold of positions (at an earlier instant in time) for the collected droplet. From this manifold and including HDI, the three equations of motion in Eq. 2.10 are advanced to determine which points on the manifold lead to a collision.

Values for the tabulated enhancement factor over the domain (10-30 μm) are presented in Figure 2.2b.

Eddy Dissipation Rates and Reynolds Numbers

In Chapter 6 stochastic parameters were acquired from the detailed evolution of the droplet spectrum at various dissipative kinetic energies and Taylor micro scale Reynolds numbers. A similar range of dissipation rates was used for both turbulent kernels. However the range of Taylor-based Reynolds numbers for each turbulent kernel differed by nearly three orders of magnitude.

The Taylor micro scale Reynolds number's (Re_λ) used by Pinsky, Khain, and collaborators (2007, 2008) were dependent on eddy dissipation rates through the use of the longitudinal Taylor micro scale as the characteristic length scale (Saffman & Turner 1956):

$$\text{Re}_\lambda = \frac{\bar{w}\lambda}{\nu} = \sqrt{15} \frac{\bar{w}^2}{(\nu\epsilon)^{0.5}} \quad (2.11)$$

where $\lambda = \bar{w} \left(\frac{15\nu}{\epsilon} \right)^{0.5}$ is the longitudinal Taylor length scale of fluid accel-

Table 2.1: Reynolds Numbers

Eddy Dissipation Rate (cm^2s^{-3})	Longitudinal Taylor-based Reynolds number	
	Franklin et. al. Parameterization	Pinsky-Khain Tables
10	27.7	5,000
50	33.6	5,000
100	36.5	10,000
200	39.7	10,000
500	44.3	20,000
1000	48.1	20,000
1500	50.5	

Table 2.1: Longitudinal Taylor-based Reynolds number are acquired from the parameterization of Franklin et. al. (2007) and from the derivations by Pinsky, Khain, and collaborators. The values in bold (red) were not given by the respective author, but are included here for convenience and perspective.

eration, $\overline{w}$ is the rms of relative velocity fluctuations, and ν is the kinematic viscosity.

This relationship between the Taylor micro scale Reynolds number and the eddy dissipation rate is in contrast to the power law parameterization of the Reynolds number by Franklin et. al. (2007):

$$\text{Re}_\lambda = 21\epsilon^{0.12}$$

Franklin’s parameterized values of the Taylor micro scale Reynolds number range from 33 to 55. The values used in Pinsky and Khain’s (2008) calculations range from 5000 to 20000. The latter range is consistent with expected values in atmospheric clouds. Both ranges correspond to eddy dissipation rates that span two orders of magnitude ($\text{cm}^2 \text{s}^{-3}$). The parameterized Reynolds number extends up to $1536 \text{ cm}^2 \text{s}^{-3}$ and the derived Reynolds number down to $10 \text{ cm}^2 \text{s}^{-3}$. The details for both methods of calculating the Reynolds numbers are presented in Table (2.1). The values highlighted in bold (red) were parameterized but have yet to be published.

2.3 Stochastic Simulation of Collisions

Gillespie's stochastic evolution algorithm (1975a) uses two independent and uniformly distributed random variables to simulate a collision. Both are dependent on the sum of the collision rates of all possible combinations of droplets. The time until the next collision is inversely proportional to the sum of the collision rates of all droplet pairs:

$$\tau = \frac{-\ln(u)}{K} \quad u \sim \mathcal{U}(0, 1), \quad K = \sum_{i < j} K_{ij}$$

where K_{ij} is compact notation for $K(x_i, x_j)$. The pair of droplets that will form a single (larger) droplet in the next collision is determined by implementing Gillespie's "first coalescence method" (Gillespie 1975a). A single uniformly selected random variable, v , is used to choose among potential source droplet-pairs (x_i, x_j) :

$$\pi_{k_0} \equiv \pi_{i_0, j_0} = \sum_{i \leq j}^{i_0, j_0} \frac{K_{ij}}{K}, \quad \pi_{k_0-1} < v \leq \pi_{k_0}, \quad v \sim \mathcal{U}(0, 1)$$

where $k \in \{1, 2, \dots, M\}$ and $M = \frac{N(N+1)}{2}$. N is the number of droplets (or bins) and $\pi_0 \equiv 0$.

Seesselberg et. al (1996) discretized the droplet size spectrum and applied Gillespie's 'pair-selection' algorithm to select the bins from which droplets are selected. They assumed a uniform intra-bin distribution of mass density and randomly selected a droplet from a subset of bin i . The interval within bin i is constructed to be an interval I_i that is symmetric about the mean intra-bin mass $\bar{x}_i$. The masses of a droplet at the left hand and right-hand boundaries of bin i are $x_{i-1/2}$ and $x_{i+1/2}$, respectively

$$I_i = [d_{i-1/2}, d_{i+1/2}] \subseteq [x_{i-1/2}, x_{i+1/2}] = \text{length of bin } i$$

Letting $\Delta m_i \equiv \min(x_{i+1/2} - \bar{x}_i, \bar{x}_i - x_{i-1/2})$, the bounds of I_i are defined by

$$d_{i-1/2} = \bar{x}_i - \Delta m_i, \quad \text{and} \quad d_{i+1/2} = \Delta m_i + \bar{x}_i$$

(Seesselberg, Trautmann, McGraw & Thorn 1996)

The initial droplet size distribution assigns some bins a number concentration $n(x, 0)$ s.t. $1 < n(x, 0) < 2$. The algorithm of Seeβelberg et. al (1996) fails to adequately restrict the interval from which the droplet-to-be-removed is selected when the number concentration is between 1 and 2. Consequently, in these situations, too much or too little mass may be removed during the next collision that selects bin i as a source bin. This results in a post-collision mean intra-bin mass that is less than the mass of the smaller bin boundary, or greater than the mass of the larger bin boundary.

The sketch in Figure (2.3) illustrates an example of when this can occur. The sketch presents one bin in the droplet size spectrum. The spectrum is described by the mass of a droplet where $\hat{g}_i$ is the total mass in bin i and

**Figure 2.3: Stochastic Droplet Selection Interval:
Seeβelberg et. al., 1996**

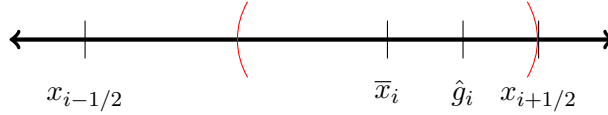


Figure 2.3: A schematic of one bin on the droplet size spectrum showing the bin boundaries for bin i , mean intra-bin mass $\bar{x}_i$, and the total amount of mass contained in bin i , $\hat{g}_i$. The red (larger) arcs are the boundaries of the interval within which a droplet is chosen for the next collision. The algorithm to determine these boundaries are by Seeβelberg et. al. (1996) and described in Section (2.3).

$\hat{g}_i = g_i \Delta x$. The mass density in bin i is g_i and the width of bin i is Δx . The two red (larger) arcs are the bounds of interval I_i and are derived in the immediately preceding paragraphs. Because the sub-interval bounded by $\hat{g}_i$ and $x_{i+1/2}$ overlaps (at least partially) the interval created by the algorithm of Seeβelberg et. al. (bounded by the red arcs), then it is possible to select an amount of mass to be removed that is greater than the total amount of mass in bin i . Also, when N_i is slightly greater than unity, say $N_i = 1 + \alpha$, with $\alpha > 0$, it is possible to select an amount of mass that is less than $\hat{g}_i$ but will result in the post-collision mean mass to be less than $x_{i-1/2}$.

A review of the literature identifies the Seeβelberg et al. (1996) paper as a reference when validating the use of bin-based moment methods such as Bott's Linear Flux Method (Pinsky and Khain 2002, and Khain et al. 2000), when identifying a Monte Carlo method (Khain et al. 2004, Bott 1998, and Sin-ichiro et al. 2009), or when quantifying a coalescence efficiency (Pinsky et al 1998, and Shin-ichiro 2009). In 1999 Trautmann, a coauthor of the

1996 article, expanded upon that paper, with other collaborators, to include an exponential in-bin distribution and log-radius coordinates (Trautmann & Wanner 1999). In 1999 Tzivion and collaborators critiqued the work of Seeßelberg et. al. (1996) by noting that it did not include sensitivity tests with respect to number of bins, kernel used, the time step, or initial conditions (Tzivion 1999). A reproduction of the bin-based modification to the stochastic simulation has not been found in the literature.

2.4 Detailed Bin-Pair Interaction Methods

Spectral methods (also known as detailed microphysics methods or Bin-pair interaction methods) are a class of numerical methods for solving the kinetic collection equation by discretizing the droplet size (mass or radius) spectrum into bins. Evolving the KCE on a discretized spectrum commonly models either the first or second moments of the droplet distribution function: number concentration and mass density, respectively. The spectrum is commonly divided such that aggregates of bins fall into two intervals: one containing cloud droplets and one containing rain droplets. When the size spectrum describes the radii of droplets, the threshold between these two segments is a droplet radius with a value between $20\mu\text{m}$ and $100\mu\text{m}$. Cloud droplets are small enough to have a terminal velocity that is dominated by typical cloud updrafts. Rain droplets are large enough that gravity dominates their velocity. With appropriate boundary conditions, the partial moments (defined in Equation 2.12): cloud mixing ratio q_c , rain mixing ratio q_r , cloud number concentration N_c , and rain number concentration N_r can be evolved (Sant et. al. 2013)

$$N_c = \int_0^{x^*} n(x)dx, \quad q_c = \int_0^{x^*} xn(x)dx, \quad N_r = \int_{x^*}^{\infty} n(x)dx, \quad q_r = \int_{x^*}^{\infty} xn(x)dx \quad (2.12)$$

where $n(x)$ is a number concentration density function with the same meaning as used in Equation 2.1.

Spectral methods discretize the droplet spectrum into bins, or categories. A logarithmic spectrum, introduced by Berry (1967) is commonly used because it provides the detail needed for the smaller particles and moderates the total number of bins by having larger bins in the downstream part of the spectrum.

Berry (1967) applied the change of variables

$$g(y, t) = 3x^2n(x, t) \quad \text{and} \quad g(y, t)dy = xn(x, t)dx$$

to Equation (2.1) where $y = \ln(r/r_0)$ and $r = (\frac{3x}{4\pi\rho_0})^{1/3}$. The radius and mass of a droplet are r and x , respectively, r_0 is the radius of the smallest droplet, and ρ_0 is the density of liquid water.

In log coordinates and as a function of radius, the kinetic collection equation describing droplet collisions is

$$\frac{\partial g(y, t)}{\partial t} = \int_{y_0}^{y_1} g(y - \acute{y}, t) \frac{x^2 K(y - \acute{y}, \acute{y})}{(x - \acute{x})^2 \acute{x}} g(\acute{y}, t) d\acute{y} - \int_{y_0}^{\infty} g(y, t) \frac{K(y, \acute{y})}{\acute{x}} g(\acute{y}, t) d\acute{y}. \quad (2.13)$$

The bounds of integration of the first integral in (2.13), y_1 and y_0 , are calculated from $x/2$ and x_0 , respectively, where x_0 is the mass of the smallest droplet. A logarithmically equidistant grid is used for the solution of (2.13) where

$$x_{k+1} = \alpha x_k, \quad k = 1, \dots, m.$$

where $\alpha = 2^{1/s}$ and for example $s = 2$ results in a doubling of the mass every two grid cells (Bott 1998).

There are three main types of detailed spectral methods: point-based methods, moment methods, and bin-pair interaction methods. Spectral methods use the kinetic collection equation to represent collision of particles and model the evolution of the particle's number and mass distribution functions.

Point-based methods represent the continuous distribution on a set of finite points, namely the mass density at each of the bin centres, and use an interpolation scheme to calculate other values of the distribution. The point-based method given by Berry and Reinhardt 1974 uses a six-point Lagrange polynomial interpolation, and the one given by Gelbart and Seinfeld (1978) uses collocation on finite elements within which the dependant variable is represented by cubic polynomials. In 1988 Eyre, Wright, and Reuter reduced the number of cubic splines in a collocation method by using an adaptive mesh. The advantages of the point-based methods are very good accuracy, no ad-hoc parameters, and the schemes are not limited to polynomial approximations of hydrodynamic kernels. However, they are not mass conservative because of the truncation error of the interpolation polynomial which is necessary when a continuous function is approximated by a

finite set of points. Berry and Reinhard’s six-point Lagrange interpolation can lead to numerical instability in the tail region (Wang 2007).

The moment methods also discretize the droplet spectrum into bins, but use moments of an adaptation of Equation (2.1) in which the bounds of integration for the loss integrals are the bin boundaries, and each gain integral is a partial moment such that one bound of integration is a bin-boundary and the other bound is an interior point of the bin. There are N sets of equations for a discretized spectrum with N bins. The moment method presented by Bleck (1970) used a mass-weighted mean-value for the number density in each bin and used applied power weighting functions to suppress the acceleration present in the spectrum evolution. Soong (1974) addressed the acceleration with exponential functions. Both Bleck’s and Soong’s methods were one moment approximations which intrinsically possess artificial acceleration (Tzivion et. al., 1987).

Enukashvily (1980) and Tzivion et. al. (1987) each present two-moment methods. The former uses a linear equation as the distribution function to approximate a partial moment for the gain integral. The latter uses a dimensionless parameter which is a function of three sequential moments within a single bin. Enukashvily (1980) assumes a constant number concentration in each bin. Using the Schwartz inequality, the higher moment can be expressed in terms of the lower two moments. These methods eliminate the need for the ad-hoc weighting function used by Bleck (1970) and by Soong (1974); and they are applicable to droplet breakup and coalescence processes. Both methods are dependant on the choice of grid points: the dimensionless parameter used by Tzivion (1999) is dependent on the bin-width as is the range of the orthogonal polynomials used by Enukashvily (1980). The application of both of these two-moment methods are limited to analytic and polynomial approximations to hydrodynamic kernels.

Bin-pair interaction methods discretize the droplet spectrum into bins and simulate collisions between all possible bin-pair combinations. In each simulated collision, mass is removed from two source bins that represent pre-collision droplets, are added together, and then deposited into target bins. The problem is similar to that of the moment methods in that the domain of the post collision mass does not coincide with the boundaries of the target bins, thus necessitating closure assumptions or approximate distribution functions. The most commonly used bin-pair interaction method is Bott’s (1998) midpoint method. It is mass conservative, computationally efficient, and not limited to polynomial approximations of hydrodynamic kernels. Bott

(1998) used a midpoint approximation to calculate the mass removed from the source bins i and j ($\dot{g}_i$ and $\dot{g}_j$):

$$\begin{aligned}\dot{g}_i &= g_i \frac{\bar{K}(i, j)}{x_j} g_j \Delta y \Delta t \\ \dot{g}_j &= g_j \frac{\bar{K}(i, j)}{x_i} g_i \Delta y \Delta t\end{aligned}$$

The pre-collision mass densities of the source bins are g_i and g_j , and the post-collision mass densities of the source bins are $g_i(i, j)$ and $g_j(i, j)$. $\bar{K}(i, j)$ is a two-dimensional interpolation of the collision kernel $K(i, j)$ explained in Bott (1998), and Δt and $\Delta y = \frac{\ln \alpha}{3}$ are the prescribed time step and bin width, respectively. The mass gained by the target bins is:

$$\dot{g}_k = \dot{g}_i + \dot{g}_j \quad (2.14)$$

The drawbacks to this method is that it is a one moment scheme that evolves mixing ratio (thus number concentration is dependent on mixing ratio), uses an ad-hoc parameter to retard excessive acceleration, and relies on target bin information to distribute post-collision mass among target bins. The bin-pair method by Simmel et. al. (2002) is a two-moment scheme and uses source-bin information to distribute post-collision mass to target bins. However, the source-bin information comes only from within the bin boundaries of the source bins. An advantage of both is neither bin-pair method is restricted to polynomial approximations of hydrodynamic kernels. The order of the kernel and number concentration approximations in the bin-pair method of Wang et. al. (2007) determines the computational expense of the Gaussian quadrature. The Gaussian quadrature gives exact solutions once the approximations are accepted. Like the method of Simmel et. al. (2002), the distribution of post-collision mass into more than one target bin uses source bin information, but that information only comes from within the two source bin boundaries.

2.5 Bulk Rate Parameterizations

Recall that self-collection (cloud and rain), auto conversion, and accretion are the collision and coalescence processes that comprise bulk rate equations. Auto conversion is the only one of these processes that appears in

each of the differential equations in the coupled set. It is also the one most strongly influenced by turbulence which makes it the most difficult to accurately model. Over the past 40+ years, more research has focused on parameterizing auto conversion, particularly the auto conversion producing rain mixing ratio, than the other processes. When a full suite of collision and coalescence parameterizations were derived, it was a common practice to make self collection and the remaining auto conversion processes to be functions of the auto conversion parameterization.

2.5.1 Auto-conversion: Rain Mixing Ratio

The seminal work for an auto conversion parameterization as a function of cloud liquid water content L_c is Kessler's work in 1969 which used a Heaviside function to terminate the auto conversion when a critical threshold was reached (Liu & Daum 2004). Liu and Daum (2004), hereafter referred to as LD04, improved the Kessler-Type parameterization by using a theoretical basis to derive a dependence on cloud droplet concentration N_c (Liu & Daum 2004). The Sunqvist-Type parameterization replaces the Heaviside function with a decaying exponential and was improved by Liu et. al. (2006), hereafter referred to as LD06, by using a similar theoretical basis to derive a dependence on cloud droplet concentration (Liu et al. 2006). Khairoutdinov and Kogan (2000), hereafter referred to as KK00, applied the least squares method to results of many simulations of a detailed microphysics method. Their input parameters spanned a state space of liquid water content and droplet concentration N . They returned a single parameterization to be used for the entire state space (Khairoutdinov & Kogan 2000). Seifert and Beheng's (2001), hereafter referred to as SB01, parameterization was derived from the kinetic collection equation. It was dependent on both L_c and the cloud droplet concentration N_c and was validated with results from a detailed microphysics method (Seifert & Beheng 2001). Franklin applied DNS results to the parameterization of Khairoutdinov and Kogan (2000) to get parameters as a function of the Taylor-based Reynolds number (Franklin 2008), hereafter referred to as Fr08, and validated the results with DNS. Franklin's turbulent parameterizations are valid for a range of turbulent kinetic energy of $100 - 1500 \text{ cm}^2\text{s}^{-3}$.

Sub-grid scale parameterizations receive input parameters such as L_c , N_c , the mixing ratio q_r , or specific humidity from climate models and return output parameters of cloud microphysical processes such as the onset of precipitation

Table 2.2: Auto-conversion Power Law

Name	ω_{au}	a	b
KK00	1350	2.47	-1.79
SB01	$f_{SB}(\kappa_c, \bar{x}_c, x^*, \tau, \nu)$	2	0
LD04	$H(\epsilon, \lambda)f_K(\kappa_c, \epsilon)$	3	-1
LL06	$\exp -\alpha^\mu f_S(\kappa_c, \epsilon)$	3	-1
Fr08, gravitation	$f_{\text{Fr,grav}}$	1.59	-1.59
Fr08, turbulent	$f_{\text{Fr,turb}}(R_\lambda)$	1.49 - 1.38	-1.35 - -1.19

Table 2.2: Power Law parameterizations for the auto-conversion process. The constants ω_{au} , a , b are taken directly from Equation (2.15). Description of each of the parameterizations are given in the text.

and the radar reflectivity. A common auto conversion parameterization, used by KK00, SB01, LD04 and LD06, and Fr08, takes a power law form:

$$\left(\frac{\partial q_r}{\partial t}\right)_{\text{au}} = \omega_{\text{au}} q_c^a N_c^b, \text{ where } \omega_{\text{au}}, a, \text{ and } b \text{ are parameters.} \quad (2.15)$$

Table 2.3: Accretion Power Law

Name	ω_{ac}	α	β
KK00	67	1.15	1.15
SB01	$g_{SB}(\kappa_r, \bar{x}_c, \bar{x}_r, x^*, \tau, \nu)$	1	1
Fr08, gravitation	4.68	1	1
Fr08, turbulent	$g_{\text{Fr}}(R_\lambda)$	1	1

Table 2.3: Power Law parameterizations of the accretion process.

Kessler's work and the work of LD04 and LD06 produced only auto-conversion parameterizations for rain mixing ratio. KK00, SB01, and Fr08 derived expressions for the auto-conversion process affecting rain mixing ratio and used

conservation of mass to deliver an expression for loss of cloud mixing ratio due to auto-conversion. They used the expression

$$\bar{x} = \frac{q}{N} \quad (2.16)$$

to derive parameterizations for the effect of auto-conversion on the cloud and rain number concentrations.

2.5.2 Accretion: Rain Mixing Ratio

As a part of their suite of bulk rate parameterizations KK00, SB01, and Fr08 separately derived expressions for the accretion process producing rain mixing ratio, and used conservation of mass and Equation (2.16) to produce related processes. A common accretion parameterization, used by KK00, SB01, and Fr08, also takes a power law form:

$$\left(\frac{\partial q_r}{\partial t} \right)_{\text{ac}} = \omega_{\text{ac}} q_c^\alpha q_r^\beta, \text{ where } \omega_{\text{ac}}, \alpha, \text{ and } \beta \text{ are parameters.}$$

In some parameterizations ω_{au} and ω_{ac} had dependencies on microphysical processes such as the collection kernel, an ad-hoc shape parameter of the gamma distribution, and arbitrary fitting parameter, or the cloud/rain droplet radius threshold (Seifert & Beheng 2001, Liu & Daum 2004, Liu et al. 2006). Franklin (2008) used the power law forms with the parameterization dependent on the Taylor-based Reynolds number. Franklin’s turbulent parameterizations are valid for a range of turbulent kinetic energy of $100 - 1500 \text{ cm}^2\text{s}^{-3}$. These parameterizations have all used a grid-box mean value for the mixing ratio. Wood et. al. (2002) showed that the use of a grid-box mean value for the mixing ratio results in an under prediction of the auto conversion rate.

2.5.3 Set of Four Coupled Equations

In each of the following parameterizations, the authors derived expressions for auto-conversion rain mixing ratio and accretion rain mixing ratio. They enforced conservation of mass by making the cloud mixing ratio expressions to be functions of the respective rain mixing ratio expressions. In each case, expressions for rain number concentration, both auto-conversion and accretion, are functions of the respective rain mixing ratio expressions, and

accuracy of number is maintained by making cloud number concentration, both auto-conversion and accretion, functions of rain number concentration. Collision and coalescence are represented by four equations that model cloud and rain self-collection, auto conversion, and accretion in the evolution of four cloud properties: cloud number, rain number, cloud mixing ratio, and rain mixing ratio (Grabowski 1999).

Franklin 2008

The non-turbulent hydrodynamic kernel Franklin (2008) used gives the following bulk rate parameterization. Franklin provided separate terms for cloud number concentration auto conversion and self-collection. The accretion and self-collection terms share the power laws of the cloud properties with the corresponding terms in the mean stochastic parameterization. The auto conversion terms, both number concentration and mass, have power laws different than the mean stochastic auto conversion term. Mass conservation is forced and auto conversion of number concentration is simply a function of mass auto conversion.

$$\begin{aligned}\frac{d N_c}{d t} &= -\frac{2}{x^*} \gamma_{\text{grav}} r_{vc}^{4.78} - \frac{4.68}{x^*} q_r N_c - (2.15 \times 10^4) q_c^2 \\ \frac{d N_r}{d t} &= \frac{\gamma_{\text{grav}}}{x^*} r_{vc}^{4.78} - 9.0 \times 10^{-3} q_r N_r \\ \frac{d q_c}{d t} &= -\gamma_{\text{grav}} r_{vc}^{4.78} - 4.68 q_c q_r \\ \frac{d q_r}{d t} &= \gamma_{\text{grav}} r_{vc}^{4.78} + 4.68 q_c q_r\end{aligned}$$

where $r_{vc} = \left(\frac{3\rho_a}{4\pi\rho_w} \frac{q_c}{N_c} \right)^{1/3}$, $\gamma_{\text{grav}} = 1.37 \times 10^{-13}$, ρ_a and ρ_w are the densities of air and water, respectively, and x^* is a threshold mass corresponding to a radius of $40\mu\text{m}$.

Franklin's turbulent parameterizations retain the cloud mass and number concentration dependencies and are a function of the Taylor-based Reynolds number as recorded from DNS of turbulent flow.

$$\begin{aligned}\frac{d N_c}{d t} &= -\frac{2}{x^*} \gamma_{\text{turb}} r_{vc}^{4.78} - \frac{\gamma_{\text{accr}}}{x^*} q_r N_c - (-2.6 \times 10^5 (0.092^{R_\lambda}) + 4.4 \times 10^4) q_c^2 \\ \frac{d N_r}{d t} &= \frac{\gamma_{\text{turb}}}{x^*} r_{vc}^{4.78} - 9.0 \times 10^{-3} q_r N_r \\ \frac{d q_c}{d t} &= -\gamma_{\text{turb}} r_{vc}^{4.78} - \gamma_{\text{accr}} q_c q_r \\ \frac{d q_r}{d t} &= \gamma_{\text{turb}} r_{vc}^{4.78} + \gamma_{\text{accr}} q_c q_r\end{aligned}$$

where $\gamma_{\text{turb}} = (2.5 \times 10^{-13} - 5.9 \times 10^{-12} \ln(R_\lambda))^{-1}$ and $\gamma_{\text{accr}} = 5.44 \exp(-3.81 R_\lambda^{-1})$.

Franklin's expression for number concentration due to rain self-collection is not affected by the turbulence in her research. Thus, the turbulent parameterization contains precisely the expression for number concentration due to rain self-collection given in the gravitational case.

Khairoutdinov and Kogan 2000

Khairoutdinov and Kogan's parameterization shares the power law for cloud accretion of number concentration, but completely omits a term for rain self-collection of number concentration. The terms for mass accretion are similar in power law to the mean stochastic mass accretion terms. As with Franklin's parameterization, mass conservation is forced and auto conversion of number concentration is simply a function of mass auto conversion.

$$\begin{aligned}\frac{dN_c}{dt} &= -1350q_c^{1.47}N_c^{-0.79} - 3.7q_rN_c \\ \frac{dN_r}{dt} &= \frac{1350}{x^*}q_c^{2.47}N_c^{-1.79} \\ \frac{dq_c}{dt} &= -1350q_c^{2.47}N_c^{-1.79} - 67(q_cq_r)^{1.15} \\ \frac{dq_r}{dt} &= 1350q_c^{2.47}N_c^{-1.79} + 67(q_cq_r)^{1.15}\end{aligned}$$

Khairoutdinov and Kogan used a threshold mass of $x^* = 25 \mu\text{m}$.

Seifert and Beheng 2001

The power laws of the cloud properties in Seifert and Beheng's parameterization closely reflect the power laws in the mean stochastic parameterization given $N_c\bar{x}_c = q_c$ and $N_r\bar{x}_r = q_r$

$$\begin{aligned}\frac{dN_c}{dt} &= \frac{\nu+2}{\nu+1}k_cq_c^2 - \frac{1}{\bar{x}_c}\Phi_{\text{ac}}(\tau)\left(1 + \frac{\nu+2}{\nu+1}\frac{\bar{x}_c}{\bar{x}_r}\right)k_rq_cq_r \\ \frac{dN_r}{dt} &= \frac{1}{20}\left(\frac{\bar{x}_c}{x^*}\right)^2\frac{(\nu+2)(\nu+4)}{(\nu+1)^2}\left[1 + \frac{\Phi_{\text{au}}}{(1-\tau)^2}\right]k_cq_c^2 - k_rq_rN_r \\ \frac{dq_c}{dt} &= -\frac{1}{20}\frac{(\bar{x}_c)^2}{x^*}\frac{(\nu+2)(\nu+4)}{(\nu+1)^2}\left[1 + \frac{\Phi_{\text{au}}}{(1-\tau)^2}\right]k_cq_c^2 - \\ &\quad \Phi_{\text{ac}}(\tau)\left(1 + \frac{\nu+2}{\nu+1}\frac{\bar{x}_c}{\bar{x}_r}\right)k_rq_cq_r \\ \frac{dq_r}{dt} &= \frac{1}{20}\frac{(\bar{x}_c)^2}{x^*}\frac{(\nu+2)(\nu+4)}{(\nu+1)^2}\left[1 + \frac{\Phi_{\text{au}}}{(1-\tau)^2}\right]k_cq_c^2 +\end{aligned}$$

$$\Phi_{\text{ac}}(\tau) \left(1 + \frac{\nu + 2 \bar{x}_c}{\nu + 1 \bar{x}_r} \right) k_r q_c q_r \quad (2.17)$$

where $k_c = 9.44 \times 10^9 \text{ cm}^3 \text{g}^{-2} \text{s}^{-1}$, and $k_r = 5.78 \times 10^3 \text{ cm}^3 \text{g}^{-1} \text{s}^{-1}$, and $0 \leq \tau \leq 1$ is non-dimensional time, and $\Phi_{\text{au}}(\tau)$ and $\Phi_{\text{ac}}(\tau)$ are fitted functions that represent internal time scales for these processes that are dependent on cloud mixing ratios. Seifert and Beheng used a threshold mass of $x^* = 40 \mu\text{m}$.

For the purpose of recovering Seifert and Beheng's (2001) parameterization, their equations are presented in a condensed form that emphasizes the dependence on the evolving partial moments. Seifert and Beheng used the centres of cloud and rain aggregates: $\bar{x}_c = q_c/N_c$ and $\bar{x}_r = q_r/N_r$ and assumed a gamma distribution as an initial condition to close the two-moment problem. They derived auto-conversion and accretion expressions as functions of their tuning parameters: a gamma distribution shape parameter, $0 \leq \nu \leq 3$, and a non-dimensional time parameter, $0 < \tau \leq 1$:

$$\begin{aligned} \Gamma_{\text{auto}} &= \frac{1}{20} \frac{(\bar{x}_c)^2}{x^*} \frac{(\nu + 2)(\nu + 4)}{(\nu + 1)^2} \left[1 + \frac{\Phi_{\text{au}}(\tau)}{(1 - \tau)^2} \right] \\ \Gamma_{\text{accr}} &= \Phi_{\text{ac}}(\tau) \left(1 + \frac{\nu + 2 \bar{x}_c}{\nu + 1 \bar{x}_r} \right). \end{aligned} \quad (2.18)$$

The specific forms of the fitted functions, $\Phi_{\text{au}}(\tau)$ and $\Phi_{\text{ac}}(\tau)$, are not relevant to the research presented herein. The details on these functions can be found in their 2001 paper (Seifert & Beheng 2001). They used a piece-wise combination of Golovin's sum of mass kernel and Long's quadratic polynomial kernel:

$$K(x, y) = \begin{cases} k_c(x^2 + y^2), & x \text{ and } y < x^* \\ k_r(x + y), & x \text{ or } y \geq x^*, \end{cases} \quad (2.19)$$

where $k_c = 9.44 \times 10^9 \text{ cm}^3 \text{g}^{-2} \text{s}^{-1}$, and $k_r = 5.78 \times 10^3 \text{ cm}^3 \text{g}^{-1} \text{s}^{-1}$. Using

abbreviated notation, their bulk model is

$$\begin{aligned}
\frac{\partial N_c}{\partial t} &= - \underbrace{\frac{\nu+2}{\nu+1} k_c q_c^2}_{\text{auto \& self-coll}} - \underbrace{\Gamma_{\text{accr}} k_r q_r N_c}_{\text{accretion}} \\
\frac{\partial N_r}{\partial t} &= \underbrace{\frac{\Gamma_{\text{auto}}}{x^*} k_c q_c^2}_{\text{autoconversion}} - \underbrace{k_r q_r N_r}_{\text{self-coll}} \\
\frac{\partial q_c}{\partial t} &= - \underbrace{\Gamma_{\text{auto}} k_c q_c^2}_{\text{autoconversion}} - \underbrace{\Gamma_{\text{accr}} k_r q_c q_r}_{\text{accretion}} \\
\frac{\partial q_r}{\partial t} &= \underbrace{\Gamma_{\text{auto}} k_c q_c^2}_{\text{autoconversion}} + \underbrace{\Gamma_{\text{accr}} k_r q_c q_r}_{\text{accretion}} .
\end{aligned} \tag{2.20}$$

The parameters ν and τ are used in seven of eight terms in their bulk parameterization. The meanings associated with each term are explained in Section 4.2.

3 | Derivation

Here, we propose a new methodology to derive bulk cloud microphysics equations from the kinetic coalescence equation in the case of warm clouds, but in principle it can be applied to more general situations. Our method is based on a systematic decomposition of the cloud and rain droplets into a mean and a set of random fluctuations. The latter leads to the closure of the high order moments as a sequence of Ornstein Uhlenbeck-like processes. In Section 3.1 we identify assumptions and approximations to the kinetic collection equation used in the stochastic bulk parameterization. Section 3.2 derives the four stochastic differential equations used in our parameterization. Section 3.3 closes the parameterization using two order of magnitude arguments on the (i) temporal fluctuations of the evolving bulk cloud properties and (ii) the relative strength of the mean and variance of the stochastic processes.

3.1 Definitions and Assumptions

For simplicity in exposition, we consider the kinetic coalescence equation (KCE) Equation 2.1 for warm clouds. Large droplets that reach certain terminal velocities escape the updraughts and fall out of the cloud as rain. The droplet spectrum is thus naturally divided into rain droplets and cloud droplets depending whether the droplet mass exceeds a heuristically chosen threshold x^* . In practice, x^* depends on the strength of the updraughts and thus it is typically larger for deep convective clouds and smaller for shallow cumulus clouds. For convenience, we assume that x^* is a fixed parameter.

Accordingly, we define the droplet numbers, N_c , N_r , and mixing ratios, q_c , q_r , of cloud (subscript ‘c’) and rain (subscript ‘r’) aggregates as follows.

$$N_c = \int_0^{x^*} n(x) dx, \quad q_c = \int_0^{x^*} xn(x) dx, \quad N_r = \int_{x^*}^{\infty} n(x) dx, \quad q_r = \int_{x^*}^{\infty} xn(x) dx \quad (3.1)$$

where $n(x)$ is a number concentration density function with the same meaning as used in Equation 2.1. We define the centres of mass of the cloud and rain aggregates as

$$\bar{x}_c \equiv \frac{1}{N_c} \int_0^{x^*} xn(x) dx = \frac{q_c}{N_c}, \quad \bar{x}_r \equiv \frac{1}{N_r} \int_{x^*}^{\infty} xn(x) dx = \frac{q_r}{N_r}. \quad (3.2)$$

The framework developed below can be easily extended to more than two such bins allowing the inclusion of drizzle, for example.

3.1.1 Density Approximations

Both number concentration density and mixing ratio density are approximated by their respective aggregate totals, normalized by the width of the spectrum on which the aggregate is defined, plus a stochastic fluctuation:

$$n(x, t) = \begin{cases} \frac{N_c(t)}{x^*} + \delta_c(\omega_1; x, t) & 0 \leq x < x^* \\ \frac{N_r(t)}{x_m - x^*} + \delta_r(\omega_1; x, t) & x^* \leq x < x_m. \\ 0 & x_m \leq x \end{cases} \quad (3.3)$$

$$q(x, t) = \begin{cases} \frac{q_c(t)}{x^*} + \gamma_c(\omega_2; x, t) & 0 \leq x < x^* \\ \frac{q_r(t)}{x_m - x^*} + \gamma_r(\omega_2; x, t) & x^* \leq x < x_m. \\ 0 & x_m \leq x \end{cases} \quad (3.4)$$

Here x_m is a large enough droplet mass so that $n(x) \approx 0$ and $q(x) \approx 0$ if $x \geq x_m$. In practice it can be taken to be the maximum possible droplet mass. The fluctuations of number concentration density $\delta_c(\omega_1; x, t)$ and $\delta_r(\omega_1; x, t)$ and mixing ratio density $\gamma_c(\omega_2; x, t)$ and $\gamma_r(\omega_2; x, t)$ are functions of random numbers ω_1 and ω_2 , have mean of zero, and are assumed to

have bounded variance (and prescribed covariance). Such stochastic fluctuations and corresponding random variables are further developed in Section 3.2. Because the first approximation is used to define the four aggregate quantities in Equation 3.1 and the two densities, number concentration and mixing ratio, given by Equations 3.3 and 3.4 respectively, are related by the physical constraint $q(x, t) = xn(x, t)$, a compatibility condition exists which is developed and resolved in the Appendix A.4.

3.1.2 2-D Domain: Source Droplet Pairs

Droplets affected by collision processes typically range in radii from $\sim 1 \mu\text{m}$ to several millimetres (Bott 1998, Wang et al. 2007, Simmel, Trautmann & Tetzlaff 2002). Droplets smaller than this range follow the streamlines of larger droplets and grow by condensation rather than by collision and coalescence, and droplets larger than this range experience breakup and rarely exceed several millimetres (Pruppacher & Klett 1997). When modelling a binary collision, the spherical water masses prior to the moment of collision are called ‘source droplets,’ and water mass in the single post-collision droplet is called the ‘target droplet.’

Two orthogonal spectra of source droplets define a two dimensional domain. The positive quadrant within this domain identifies all possible pre-collision droplet pair combinations as shown in Figure 3.1. The separation between rain and cloud droplets on this graph is indicated by a separation mass x^* . The mass of the separation threshold x^* between rain and cloud droplets and the largest droplet mass x_m , are indicated on the graph. In subsequent subsections, the kinetic collection equation is integrated using x_m in upper integration bounds and $x_m \rightarrow \infty$.

The shape of the domain identified in Figure 3.1 is produced from a change of variables:

$$z = x - x' \quad \text{and} \quad y = x' \quad (3.5)$$

where the original two variables ($x - x'$ and x') represent the two source droplets in the first integral in Equation 2.1. The change of variables is helpful to integrate the partial moment on the rain portion of the spectrum. The distinction between regions Ω_5 and Ω_6 facilitates the integration. Both the lower and upper bounds of integration in region Ω_6 contain x_m which is taken in the limit to infinity. Therefore all results in this region will be zero. However, the calculations are detailed in the derivations for completeness.

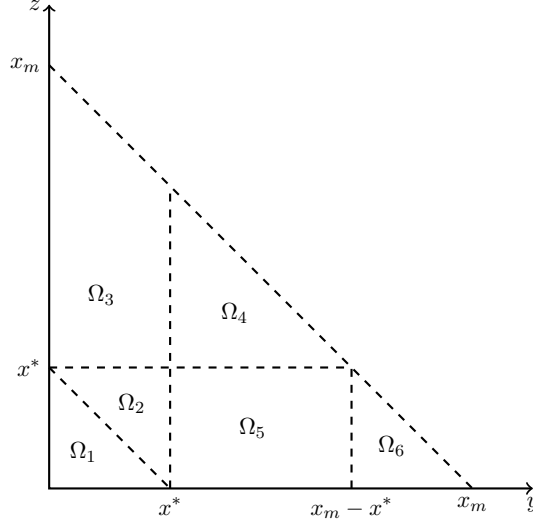
Figure 3.1: Bulk Partitions for Source Droplet Pairs

Figure 3.1: The domain of pre-collision droplet pairs is partitioned into six regions defining the collision and coalescence processes: cloud self-collection (Ω_1), auto-conversion (Ω_2), accretion (Ω_3 , Ω_5 , Ω_6), and rain self-collection (Ω_4).

The pre-collision droplet pair combinations are partitioned in Figure 3.1 into four aggregates: cloud-cloud ($\Omega_1 \cup \Omega_2$), rain-cloud (Ω_3), cloud-rain ($\Omega_5 \cup \Omega_6$), and rain-rain (Ω_4). The collision-coalescence processes are represented as follows: cloud self-collection (Ω_1), auto-conversion (Ω_2), accretion (Ω_3 , Ω_5 , Ω_6), and rain self-collection (Ω_4). The three regions given by Ω_3 , Ω_4 , and $\Omega_5 \cup \Omega_6$ each span a complete aggregate and are each represented by a single collision-coalescence process. Therefore, the integral of the fluctuations for each of these regions is zero. In Section 3.2 we show that the the state space mean of the fluctuations is zero for a complete aggregate.

The bounds of integration for the six regions in Figure 3.1 are explicitly

detailed as follows:

$$\begin{aligned}
\int_{\Omega_1} d\Omega_1 &\equiv \int_0^{x^*} \int_0^{x^*-y} dz dy && \text{cloud self-collection} \\
\int_{\Omega_2} d\Omega_2 &\equiv \int_0^{x^*} \int_{x^*-y}^{x^*} dz dy && \text{auto-conversion} \\
\int_{\Omega_3} d\Omega_3 &\equiv \int_0^{x^*} \int_{x^*}^{x_m-y} dz dy && \text{accretion} \\
\int_{\Omega_4} d\Omega_4 &\equiv \int_{x^*}^{x_m-x^*} \int_{x^*}^{x_m-y} dz dy && \text{rain self-collection} \\
\int_{\Omega_5} d\Omega_5 &\equiv \int_{x^*}^{x_m-x^*} \int_0^{x^*} dz dy && \text{accretion} \\
\int_{\Omega_6} d\Omega_6 &\equiv \int_{x_m-x^*}^{x_m} \int_0^{x_m-y} dz dy && \text{accretion}
\end{aligned} \tag{3.6}$$

In 2007 Wang and collaborators presented a figure (their Figure 2) constructed with pairs of source droplets. However, they identified bin indices on a pair of discretized spectra and omitted references to the types of collision processes associated with the distinctive regions of the domain. Figure 3.1 replaces the bin indices with physically relevant collision processes.

3.1.3 Collision Kernel Expansion

A Taylor approximation of $K(x, x')$ is centred around $\bar{x}_c$ and $\bar{x}_r$ to obtain closed forms for the bulk equations to be developed in Section 3.2.

$$\begin{aligned}
 K(x, x') = K(\bar{x}, \bar{x}') &+ \frac{\partial K(\bar{x}, \bar{x}')}{\partial x}(x - \bar{x}) + \frac{\partial K(\bar{x}, \bar{x}')}{\partial x'}(x' - \bar{x}') + \\
 &\frac{\partial^2 K(\bar{x}, \bar{x}')}{\partial x \partial x'}(x - \bar{x})(x' - \bar{x}') + \frac{\partial^2 K(\bar{x}, \bar{x}')}{2 \partial x^2}(x - \bar{x})^2 + \\
 &\frac{\partial^2 K(\bar{x}, \bar{x}')}{2 \partial x'^2}(x' - \bar{x}')^2 + \text{h.o.t},
 \end{aligned} \tag{3.7}$$

More precisely we have bins with $a \leq x, \bar{x} < b$ and $a' \leq x', \bar{x}' < b'$ s.t. $\bar{x} = \bar{x}_c$ ($\bar{x}' = \bar{x}_c$) if $a = 0$ and $b = x^*$ ($a' = 0$ and $b' = x^*$), and $\bar{x} = \bar{x}_r$ ($\bar{x}' = \bar{x}_r$) if $a = x^*$ and $b = \infty$ ($a' = x^*$ and $b' = \infty$).

The first six terms of the Taylor approximation yield a combination of constant, linear, and product kernels considered in Drake's paper (Drake & Wright 1972) to derive benchmark exact solutions for the coalescence equation. A combination of those benchmark solutions could be used here to close the equations without any further approximation of $n(x, t)$. However, that route increases the number of terms in the resultant bulk parameterization five-fold. By using only the constant term in the Taylor approximation the number of terms and associated computational costs mirror that of existing bulk microphysical parameterizations. However, unlike existing parameterizations, which are restricted to a chosen collision kernel applied during the derivation of the kinetic collection equation, the stochastic bulk parameterization can accept arbitrary collision kernels without any further derivations. We consider the stochastic correction terms to be in some way representative of the Taylor error terms.

3.2 Stochastic Bulk Model

The density approximations given by Equations 3.3 and 3.4 are used when taking moments of Equation 3.1. By construction, the instantaneous means, with respect to droplet size, of the stochastic fluctuations are zero. The sum

of the density approximations over an entire region is zero:

$$N_c(t) = \int_0^{x^*} n(x, t) dx = \int_0^{x^*} \frac{N_c(t)}{x^*} + \delta_c(\omega_1; x, t) dx = N_c(t) + \int_0^{x^*} \delta_c(\omega_1; x, t) dx$$

Consequently,

$$0 = \frac{1}{x^*} \int_0^{x^*} \delta_c(x) dx \equiv \langle \delta_c(x) \rangle, \quad (3.8)$$

where only the droplet mass dependency is expressed to emphasize that the mean is taken over an interval in the droplet spectrum and at an instant in time. Similarly, we have

$$\begin{aligned} \int_{x^*}^{x_m} \delta_r(x) dx &\equiv \langle \delta_r(x) \rangle = 0, & \int_0^{x^*} \gamma_c(x) dx &\equiv \langle \gamma_c(x) \rangle = 0, \text{ and} \\ \int_{x^*}^{x_m} \gamma_r(x) dx &\equiv \langle \gamma_r(x) \rangle = 0 \text{ for all } t. \end{aligned} \quad (3.9)$$

Moreover, we assume that δ_c and δ_r are spatially correlated with non-trivial covariance functions. Using the change of variables from Equation 3.5 for the pre-collision droplet masses, the product fluctuation terms can be expressed as:

$$\phi_{jk} = \int_{\Omega_i} \delta_j(\cdot) \delta_k(\cdot) d\Omega_i, \quad \text{or} \quad \phi_{jk} = \int_{\Omega_i} \delta_j(\cdot) \gamma_k(\cdot) d\Omega_i \quad (3.10)$$

where Ω_i is a domain identified in Figure 3.1 and $(j, k \in \{c, r\})$, and the fluctuations are a function of either z or y and are thus represented as, for example, $\delta_k(\cdot)$.

The stochastic terms are discussed using the change of variables in Equation 3.5. The symmetry of Ω_1 , with respect to the line $z = y$, eliminates the need to distinguish between the integration variables in the indexing scheme. There are two stochastic processes associated with number concentration fluctuations; one with respect to ‘ z ’ and the other with respect to ‘ y .’ Due to the symmetry of Ω_1 , they are equivalent and are combined in the second term on the right-hand-side of Equation 3.14. No further identification of fluctuations is necessary for the indexing of processes within this differential

equation. The change of variables in Equation 3.5 is used to define the following stochastic processes:

As the cloud properties evolve, the aggregate fluctuations and aggregate product fluctuations change. Consider the evolution of these instantaneous statistics to be a stochastic process:

$$X_s(t) \equiv \begin{cases} \frac{\kappa}{\Phi} \int_{\Omega_i} \delta_j(\cdot, t) d\Omega_i & \text{sum, at an instant, of a set of} \\ & \text{number concentration fluctuations} \\ \frac{\kappa}{\Phi} \int_{\Omega_i} \gamma_j(\cdot, t) d\Omega_i & \text{sum, at an instant, of a set of} \\ & \text{mixing ratio fluctuations} \\ \frac{1}{\Phi} \phi_{jk}(t) & \text{instantaneous product fluctuation,} \end{cases}$$

where $s = s(i, j, k, \Phi)$ is an indexing scheme to be developed in the following subsections, κ is a normalization constant from Equations (3.3 and 3.4), and Φ is an appropriate function of cloud properties (N_c, N_r, q_c, q_r) which serves to non-dimensionalize the stochastic process. The time dependence is reintroduced in the number concentration and mixing ratio fluctuations to show the dependence of $X_s(t)$ on time. We assume that the stochastic processes $X_s(t)$ are time homogeneous with a fixed mean μ_s and variance σ_s^2 . We thus set

$$X_s(t) = \mu_s + \sigma_s \xi_s(t), \quad (3.11)$$

where $\xi_s(t)$ is a homogeneous process with mean zero and variance one.

3.2.1 Cloud Number Concentration

The evolution of cloud number concentration is expressed as a partial moment of the kinetic collection equation over the cloud droplet portion of the spectrum.

$$\begin{aligned} \frac{dN_c}{dt} = \frac{1}{2} \int_0^{x^*} \int_0^x n(x-x')n(x')K(x-x', x')dx'dx - \\ \int_0^{x^*} \int_0^\infty n(x)n(x')K(x, x')dx'dx. \end{aligned}$$

The substitutions for the number concentration density are used, and kernel values at the mean mass of each partition requires splitting the second double integral into two terms:

$$\begin{aligned} \frac{dN_c}{dt} = & \frac{1}{2} \int_0^{x^*} \int_0^x \left[\frac{N_c}{x^*} + \delta_c(x - x') \right] \left[\frac{N_c}{x^*} + \delta_c(x') \right] K(\bar{x}_c, \bar{x}_c) dx' dx \\ & - \int_0^{x^*} \int_0^{x^*} n(x)n(x')K(\bar{x}_c, \bar{x}_c)dx'dx - \int_0^{x^*} \int_{x^*}^{\infty} n(x)n(x')K(\bar{x}_c, \bar{x}_r)dx'dx. \end{aligned} \quad (3.12)$$

Expanding the substitutions for number concentration density gives three terms in the first integral that contain stochastic fluctuations. An abbreviated notation is used for the auto conversion/cloud self-collection and accretion kernels: $K_{cc} = K(\bar{x}_c, \bar{x}_c)$ and $K_{cr} = K(\bar{x}_c, \bar{x}_r)$, respectively.

$$\begin{aligned} \frac{dN_c}{dt} = & \frac{K_{cc}}{2} \int_0^{x^*} \int_0^x \frac{N_c^2}{(x^*)^2} + \frac{N_c}{x^*} \delta_c(x - x') + \frac{N_c}{x^*} \delta_c(x') + \delta_c(x - x')\delta_c(x') dx' dx \\ & - K_{cc} \int_0^{x^*} \int_0^{x^*} n(x)n(x')dx'dx - K_{cr} \int_0^{x^*} \int_{x^*}^{\infty} n(x)n(x')dx'dx. \end{aligned} \quad (3.13)$$

The first term in the first integral consists of only constants and thus can be integrated. The second and third terms in the first integral contain stochastic fluctuations which can be integrated analytically. We will detail the term containing $\delta_c(x')$:

$$\begin{aligned} \frac{K_{cc}N_c}{2x^*} \int_0^{x^*} \int_0^x \delta_c(x')dx'dx &= \frac{K_{cc}N_c}{2x^*} \int_0^{x^*} \int_0^x \left(n(x') - \frac{N_c}{x^*} \right) dx'dx = \\ & \frac{K_{cc}N_c}{2x^*} \int_0^{x^*} n(x') \int_{x'}^{x^*} dx dx' - \frac{K_{cc}N_c^2}{4} = \\ & \frac{K_{cc}N_c}{2} \int_0^{x^*} n(x')dx' - \frac{K_{cc}N_c}{2x^*} \int_0^{x^*} x' n(x')dx' - \frac{K_{cc}N_c^2}{4} = \\ \frac{K_{cc}N_c^2}{2} - \frac{K_{cc}N_c}{2x^*} q_c - \frac{K_{cc}N_c^2}{4} &= \frac{K_{cc}N_c^2}{4} - \frac{K_{cc}N_c^2}{2x^*} \frac{q_c}{N_c} = \frac{K_{cc}N_c^2}{4} - \frac{K_{cc}N_c^2}{2} \frac{\bar{x}_c}{x^*} \end{aligned}$$

By symmetry of the region Ω_1 , the details solving the other term are similar and with a change of variables leads to the same result.

$$\begin{aligned} & \frac{K_{cc}N_c}{2x^*} \int_0^{x^*} \int_0^x \delta_c(x') dx' dx + \frac{K_{cc}N_c}{2x^*} \int_0^{x^*} \int_0^x \delta_c(x-x') dx' dx = \\ & \left(\frac{1}{4} - \frac{\bar{x}_c}{2x^*} \right) K_{cc}N_c^2 + \left(\frac{1}{4} - \frac{\bar{x}_c}{2x^*} \right) K_{cc}N_c^2 = \left(\frac{1}{2} - \frac{\bar{x}_c}{x^*} \right) K_{cc}N_c^2. \end{aligned}$$

However, this constraint limits the usability of the two-moment bulk equations because it leads to unphysical conditions that violate first principles such as conservation of mass and consistency of number concentration. Here, we relax this constraint and assume that this term is represented through a random process, denoted below by X_{1nf} . This relaxation is justified, for instance, by the severe truncation applied to the kernel function in Equation 3.7.

The second and third integrals in Equation 3.13 are partial moments over complete partitions of the droplet size spectrum and thus can also be integrated to obtain $K_{cc}N_c^2$ and $K_{cr}N_cN_r$, respectively.

The double integrals of the terms containing fluctuations and product fluctuations are aggregate fluctuations over a region at an instant. They are non-dimensionalized and represented by the stochastic processes X_s . Combining the analytically integrated terms with the stochastic process terms yields a stochastic differential equation for the evolution of cloud number concentration:

$$\frac{dN_c}{dt} = \frac{1}{4}K_{cc}N_c^2 + K_{cc}N_c^2X_{1nf} + \frac{1}{2}K_{cc}N_c^2X_{1np} - K_{cc}N_c^2 - K_{cr}N_cN_r \quad (3.14)$$

The number in the indices of these stochastic processes, X_s , identify the domain according to Figure 3.1. The first Latin letter, ‘ n ’ or ‘ m ,’ indicates number concentration or mass density, respectively. The second Latin letter, ‘ f ’ or ‘ p ,’ indicates fluctuation or product fluctuation, respectively.

Because of the symmetry of region Ω_1 the first two drift terms are identical. As already anticipated, we set

$$(\mu_{1nf} + \sigma_{1nf}\xi_{1nf}) = X_{1nf} \equiv \frac{1}{x^*N_c} \int_{\Omega_1} \delta_c(\cdot) d\Omega_1$$

$$\begin{aligned}
&= \frac{1}{2x^*N_c} \int_{\Omega_1} \delta_c(z) d\Omega_1 + \frac{1}{2x^*N_c} \int_{\Omega_1} \delta_c(y) d\Omega_1, \\
(\mu_{1np} + \sigma_{1np}\xi_{1np}) &= X_{1np} \equiv \frac{1}{N_c^2} \int_{\Omega_1} \delta_c(z) \delta_c(y) d\Omega_1,
\end{aligned} \tag{3.15}$$

where ξ_{1nf} and ξ_{1np} are homogeneous processes with mean zero and variance one, and μ and σ are the ensemble mean and standard deviation, respectively. Substituting Equations 3.15 into Equation 3.14, and collecting like terms gives

$$\begin{aligned}
\frac{dN_c}{dt} &= - \left(\frac{3}{4} - \frac{2\mu_{1nf} + \mu_{1np}}{2} \right) K_{cc} N_c^2 - K_{cr} N_c N_r + \\
&\quad \frac{1}{2} [2\sigma_{1nf}\xi_{1nf} + \sigma_{1np}\xi_{1np}] K_{cc} N_c^2.
\end{aligned} \tag{3.16}$$

This is the stochastic differential equation for cloud aggregate number concentration, which contains the means and variances of stochastic processes as physically meaningful parameters.

3.2.2 Rain Number Concentration

The evolution of rain number concentration is also expressed as a partial moment of the kinetic collection equation.

$$\begin{aligned}
\frac{dN_r}{dt} &= \frac{1}{2} \int_{x^*}^{\infty} \int_0^x n(x-x')n(x')K(x-x',x')dx'dx - \\
&\quad \int_{x^*}^{\infty} \int_0^{\infty} n(x)n(x')K(x,x')dx'dx
\end{aligned} \tag{3.17}$$

The convenient change of variables given in Equation 3.5 and the substitutions for the kernel allow the first integral to be partitioned into five integrals:

$$\begin{aligned}
\frac{1}{2} \int_{x^*}^{\infty} \int_0^x n(x-x')n(x')K(x-x',x')dx'dx &= \frac{K(\bar{x}_c, \bar{x}_c)}{2} \int_{\Omega_2} n(y)n(z) d\Omega_2 + \\
\frac{K(\bar{x}_c, \bar{x}_r)}{2} \left[\int_{\Omega_3} + \int_{\Omega_5} + \int_{\Omega_6} \right] n(y)n(z) d\Omega &+ \frac{K(\bar{x}_r, \bar{x}_r)}{2} \int_{\Omega_4} n(y)n(z) d\Omega_4
\end{aligned} \tag{3.18}$$

The accretion terms, Ω_3 , Ω_5 , and Ω_6 , integrate analytically to $1/2$, $1/2$, and zero, respectively. The rain self-collection term integrates to $1/2K(\bar{x}_r, \bar{x}_r)N_r^2$.

The truncation of a variety of turbulent and non-turbulent kernels introduces uncertainty into an otherwise deterministic expression. The density approximations in Equations 3.3 and 3.4 can restore the uncertainty to these terms. However, data from detailed simulations show that the parameter values associated with accretion are $\sim O(10^{-19}) - O(10^{-21})$ while those associated with rain self-collection are $\sim O(10^{-1}) - O(10^{-2})$. Therefore, we analytically integrate the accretion terms and use stochastic processes to represent the uncertainty associated with kernel truncation in the rain self-collection term. Using the substitutions for number concentration, the stochastic terms contained in the first and third terms on the right hand side of Equation 3.18 are:

$$\begin{aligned} & \frac{K(\bar{x}_c, \bar{x}_c)}{2} \int_{\Omega_2} \left[\frac{N_c}{x^*} + \delta_c(z) \right] \left[\frac{N_c}{x^*} + \delta_c(y) \right] d\Omega_2, \\ & \frac{K(\bar{x}_r, \bar{x}_r)}{2} \lim_{x_m \rightarrow \infty} \int_{\Omega_4} \left[\frac{N_r}{x_m - x^*} + \delta_r(z) \right] \left[\frac{N_r}{x_m - x^*} + \delta_r(y) \right] d\Omega_4. \end{aligned} \quad (3.19)$$

The second term in Equation 3.17 partitions into two integrals and integrates easily to yield:

$$K_{cr}N_cN_r + K_{rr}N_r^2. \quad (3.20)$$

Expanding the substitutions for number concentration density in Equation 3.19, combining that with Equation 3.20, and recognizing that the accretion terms cancel ($1/2+1/2-1=0$) produces an equation with 4 stochastic processes:

$$\begin{aligned} \frac{dN_r}{dt} = & \frac{K_{cc}}{2} \left(\frac{1}{2}N_c^2 + 2N_c^2X_{2nf} + N_c^2X_{2np} \right) + \\ & \frac{K_{rr}}{2} \lim_{x_m \rightarrow \infty} \left(\frac{(x_m - 2x^*)^2}{2(x_m - x^*)^2} N_r^2 + 2N_r^2X_{4nf} + N_r^2X_{4np} \right) - K_{rr}N_r^2 \end{aligned} \quad (3.21)$$

where $K_{cc} = K(\bar{x}_c, \bar{x}_c)$, $K_{cr} = K(\bar{x}_c, \bar{x}_r)$, and $K_{rr} = K(\bar{x}_r, \bar{x}_r)$. As anticipated X_{2nf} , X_{2np} , X_{4nf} , X_{4np} are stochastic processes with their own means

and variances, satisfying:

$$\begin{aligned}
(\mu_{2nf} + \sigma_{2nf}\xi_{2nf}) &= X_{2nf} \equiv \frac{1}{x^* N_c} \int_{\Omega_2} \delta_c(\cdot) d\Omega_2, \\
(\mu_{2np} + \sigma_{2np}\xi_{2np}) &= X_{2np} \equiv \frac{1}{N_c^2} \int_{\Omega_2} \delta_c(\cdot) \delta_c(\cdot) d\Omega_2, \\
(\mu_{4nf} + \sigma_{4nf}\xi_{4nf}) &= X_{4nf} \equiv \lim_{x_m \rightarrow \infty} \frac{1}{(x_m - x^*) N_r} \int_{\Omega_4} \delta_r(\cdot) d\Omega_4, \\
(\mu_{4np} + \sigma_{4np}\xi_{4np}) &= X_{4np} \equiv \lim_{x_m \rightarrow \infty} \frac{1}{N_r^2} \int_{\Omega_4} \delta_r(\cdot) \delta_r(\cdot) d\Omega_4,
\end{aligned} \tag{3.22}$$

where again ξ_{2nf} , ξ_{2np} , and ξ_{4np} are homogeneous processes with mean zero and variance one. When $x_m \rightarrow \infty$, we obtain $\overline{X_{4nf}} = 0$. Taking the limit in Equation 3.21, substituting the three remaining means and variances, and collecting like terms gives the stochastic parameterization for rain number concentration:

$$\begin{aligned}
\frac{d N_r}{dt} &= \left(\frac{1}{4} + \frac{2\mu_{2nf} + \mu_{2np}}{2} \right) K_{cc} N_c^2 - \left(\frac{3}{4} - \mu_{4np} \right) K_{rr} N_r^2 + \\
&\quad \frac{1}{2} (2\sigma_{2nf}\xi_{2nf} + \sigma_{2np}\xi_{2np}) K_{cc} N_c^2 + \sigma_{4np}\xi_{4np} K_{rr} N_r^2
\end{aligned} \tag{3.23}$$

This is the differential equation for rain aggregate number concentration, which contains the means of stochastic processes as physically meaningful parameters.

We note that the analytic and stochastic rain collection terms, $0.5 K_{rr} N_r^2$ and $(0.75 - \mu_{4np}) K_{rr} N_r^2$ respectively, are equal when $\mu_{4np} = 0.25$. Results from detailed simulations produce values of μ_{4np} near 0.25 that vary depending on the kernel and the age of the cloud, as will be reported in Chapters 5 and 6.

3.2.3 Cloud Mixing Ratio

The evolution of cloud mixing ratio is expressed as a partial moment of the kinetic collection equation over the cloud droplet portion of the spec-

trum.

$$\begin{aligned} \frac{dq_c}{dt} = & \frac{1}{2} \int_0^{x^*} \int_0^x x n(x-x') n(x') K(x-x', x') dx' dx - \\ & \int_0^{x^*} \int_0^\infty x n(x) n(x') K(x, x') dx' dx \end{aligned}$$

Using the substitutions for the number concentration density, mixing ratio, and the collision kernel, and applying the change of variables gives

$$\begin{aligned} \frac{dq_c}{dt} = & \frac{1}{2} \int_0^{x^*} \int_0^{x^*-y} \left[\frac{q_c}{x^*} + \gamma_c(z) \right] \left[\frac{N_c}{x^*} + \delta_c(y) \right] K(\bar{x}_c, \bar{x}_c) dz dy \\ & + \frac{1}{2} \int_0^{x^*} \int_0^{x^*-y} \left[\frac{N_c}{x^*} + \delta_c(z) \right] \left[\frac{q_c}{x^*} + \gamma_c(y) \right] K(\bar{x}_c, \bar{x}_c) dz dy \\ & - \int_0^{x^*} \int_0^{x^*} x n(x) n(x') K(\bar{x}_c, \bar{x}_c) dx' dx - \int_0^{x^*} \int_{x^*}^\infty x n(x) n(x') K(\bar{x}_c, \bar{x}_r) dx' dx \end{aligned} \quad (3.24)$$

Expanding the substitutions for number concentration density and mixing ratio density produces three terms that contain stochastic fluctuations in each of the first two integrals. The abbreviated notation for the kernels are used: $K_{cc} = K(\bar{x}_c, \bar{x}_c)$ and $K_{cr} = K(\bar{x}_c, \bar{x}_r)$.

$$\begin{aligned} \frac{dq_c}{dt} = & \frac{K_{cc}}{2} \int_0^{x^*} \int_0^{x^*-y} \frac{q_c N_c}{(x^*)^2} + \frac{q_c}{x^*} \delta_c(y) + \frac{N_c}{x^*} \gamma_c(z) + \gamma_c(z) \delta_c(y) dz dy \\ & + \frac{K_{cc}}{2} \int_0^{x^*} \int_0^{x^*-y} \frac{q_c N_c}{(x^*)^2} + \frac{q_c}{x^*} \delta_c(z) + \frac{N_c}{x^*} \gamma_c(y) + \gamma_c(y) \delta_c(z) dz dy \quad (3.25) \\ & - K_{cc} \int_0^{x^*} \int_0^{x^*} x n(x) n(x') dx' dx - K_{cr} \int_0^{x^*} \int_{x^*}^\infty x n(x) n(x') dx' dx \end{aligned}$$

The first term in each of the first two integrals contain only constants and thus can be integrated.

$$\frac{K_{cc}}{2} \int_0^{x^*} \int_0^{x^*-y} \frac{q_c N_c}{(x^*)^2} dz dy = \frac{K_{cc} q_c N_c}{2(x^*)^2} \int_0^{x^*} (x^* - y) dz dy = \frac{K_{cc} q_c N_c}{2(x^*)^2} \frac{(x^*)^2}{2} \quad (3.26)$$

Adding the first terms on the right hand side in the first two lines of Equation 3.25 gives the first term on the right hand side of Equation 3.29. The second terms in these first two integrals are the same, except for a different argument, as the ones exposed in the derivation of cloud number concentration in Section 3.2.1.

$$\frac{K_{cc} q_c}{2x^*} \int_0^{x^*} \int_0^{x^*-y} \delta_c(y) dz dy = \frac{K_{cc} q_c N_c}{2} \left(\frac{1}{x^* N_c} \int_0^{x^*} \int_0^{x^*-y} \delta_c(y) dz dy \right) \quad (3.27)$$

The term in parenthesis in Equation 3.27 is precisely X_{1nf} which is detailed in Equation 3.15. There are two occurrences of this term in Equation 3.25, and they are added to give $K_{cc} q_c N_c X_{1nf}$. These terms are also represented here as the drift component of an Ornstein-Uhlenbeck-like process. The third terms contains γ_c , which produce second moments, and yield a closure problem.

$$\frac{1}{x^* q_c} \int_{\Omega_1} \gamma_c(z) d\Omega_1 = \frac{1}{2} - \frac{1}{q_c x^*} \int_0^{x^*} z^2 n(z) dz \quad (3.28)$$

The third and fourth integrals are partial moments over complete regions and thus can be integrated. Combining the analytically integrated terms with stochastic process terms yields a stochastic differential equation for the evolution of cloud number concentration:

$$\begin{aligned} \frac{dq_c}{dt} = & \frac{1}{2} K_{cc} q_c N_c + K_{cc} q_c N_c X_{1nf} + K_{cc} q_c N_c X_{1mf} + K_{cc} q_c N_c X_{1mp} \\ & - K_{cc} q_c N_c - K_{cr} q_c N_r \end{aligned} \quad (3.29)$$

where X_{1nf} is detailed in Section 3.2.1, and

$$\begin{aligned} (\mu_{1mf} + \sigma_{1mf} \xi_{1mf}) = X_{1mf} & \equiv \frac{1}{x^* q_c} \int_{\Omega_1} \gamma_c(\cdot) d\Omega_1, \\ (\mu_{1mp} + \sigma_{1mp} \xi_{1mp}) = X_{1mp} & \equiv \frac{1}{q_c N_c} \int_{\Omega_1} \delta_c(\cdot) \gamma_c(\cdot) d\Omega_1, \end{aligned} \quad (3.30)$$

with as usual ξ_{1mf} and ξ_{1mp} being homogeneous processes with mean zero and variance one. The presence of γ in X_{1mp} yields another third moment, the closure of which is handled similarly to X_{1mf} .

The change of variables given in Equation 3.5 produces two of each of the fluctuation integrals in region Ω_1 which are summed to produce the two stochastic processes in Equation 3.30. This is done in a similar fashion as X_{1nf} in Equation 3.15. The symmetry in region Ω_1 means that the duplicate processes are equivalent; thus, the coefficient in each positive term on the rhs of Equation 3.29 is twice the coefficient in the corresponding term in Equation 3.14. The sum of the second and third terms in Equation 3.29 correspond to the second term in Equation 3.14 because they are all ‘fluctuation’ terms. Substituting Equation 3.30 in to Equation 3.29 and collecting like terms gives

$$\begin{aligned} \frac{dq_c}{dt} = & - \left(\frac{1}{2} - (\mu_{1nf} + \mu_{1mf} + \mu_{1mp}) \right) K_{cc} q_c N_c - K_{cr} q_c N_r + \\ & \frac{1}{2} [\sigma_{1nf} \xi_{1nf} + \sigma_{1mf} \xi_{1mf} + \sigma_{1mp} \xi_{1mp}] K_{cc} q_c N_c. \end{aligned} \quad (3.31)$$

This is the differential equation for cloud aggregate mixing ratio, which contains the means of stochastic processes as physically meaningful parameters.

3.2.4 Rain Mixing Ratio

The evolution of rain mixing ratio is also expressed as a partial moment of the kinetic collection equation; however, collisions with cloud droplets are included.

$$\begin{aligned} \frac{dq_r}{dt} = & \frac{1}{2} \int_{x^*}^{\infty} \int_0^x x n(x-x') n(x') K(x-x', x') dx' dx \\ & - \int_{x^*}^{\infty} \int_0^{\infty} x n(x) n(x') K(x, x') dx' dx. \end{aligned} \quad (3.32)$$

The convenient change of variables given in Equation 3.5 and the substitutions for the kernel allow the first integral to be partitioned into five inte-

grals:

$$\begin{aligned}
\frac{1}{2} \int_{x^*}^{\infty} \int_0^x n(x-x')n(x')K(x-x',x')dx'dx = & \\
\frac{K(\bar{x}_c, \bar{x}_c)}{2} \int_{\Omega_2} q(y)n(z) + n(y)q(z) d\Omega_2 + & \\
\frac{K(\bar{x}_c, \bar{x}_r)}{2} \left[\int_{\Omega_3} + \int_{\Omega_5} + \int_{\Omega_6} \right] q(y)n(z) d\Omega + & \quad (3.33) \\
\frac{K(\bar{x}_c, \bar{x}_r)}{2} \left[\int_{\Omega_3} + \int_{\Omega_5} + \int_{\Omega_6} \right] n(y)q(z) d\Omega + & \\
\frac{K(\bar{x}_r, \bar{x}_r)}{2} \int_{\Omega_4} q(y)n(z) + n(y)q(z) d\Omega_4 &
\end{aligned}$$

The coefficients in each set of accretion terms, corresponding to Ω_3 , Ω_5 , and Ω_6 , integrate analytically to $1/2$, $1/2$, and zero, respectively. The rain self-collection term integrates to $K(\bar{x}_r, \bar{x}_r)N_r^2$. Using the substitutions for number concentration and mixing ratio, the remaining terms contain stochastic fluctuations:

$$\frac{K(\bar{x}_c, \bar{x}_c)}{2} \int_{\Omega_2} \left(\left[\frac{q_c}{x^*} + \gamma_c(z) \right] \left[\frac{N_c}{x^*} + \delta_c(y) \right] + \left[\frac{q_c}{x^*} + \gamma_c(y) \right] \left[\frac{N_c}{x^*} + \delta_c(z) \right] \right) d\Omega_2 \quad (3.34)$$

The second term in Equation 3.32 partitions into two integrals and integrates easily to yield:

$$K_{cr}q_rN_c + K_{rr}q_rN_r \quad (3.35)$$

which conveniently cancel with the q_rN_c accretion terms and the rain self-collection term.

The substitutions for number concentration density and mixing ratio density are expanded. The accretion terms containing q_rN_c cancel: $(1/2+1/2-1=0)$ while the accretion terms with q_cN_r sum to unity. Combining Equations (3.34 & 3.35) produces an equation with 2 new stochastic processes:

$$\frac{dq_r}{dt} = \frac{K_{cc}}{2} (q_cN_c + 2q_cN_cX_{2nf} + 2N_cq_cX_{2mf} + 2q_cN_cX_{2mp}) + K_{cr}q_cN_r \quad (3.36)$$

where X_{2nf} is detailed in Section 3.2.3, and the remaining means and variances of the stochastic processes in Equation 3.36 are defined as:

$$\begin{aligned} (\mu_{2mf} + \sigma_{2mf}\xi_{2mf}) = X_{2mf} &\equiv \frac{1}{x^*q_c} \int_{\Omega_2} \gamma_c(\cdot) d\Omega_2, \\ (\mu_{2mp} + \sigma_{2mp}\xi_{2mp}) = X_{2mp} &\equiv \frac{1}{q_c N_c} \int_{\Omega_2} \gamma_c(\cdot) \delta_c(\cdot) d\Omega_2, \end{aligned} \quad (3.37)$$

and ξ_{2mf} and ξ_{2mp} are homogeneous processes with mean zero and variance one. The third moment that arises in X_{2mf} is the additive inverse of the expression for the third moment in X_{1nf} as shown in the right hand side of Equation 3.28, and similarly X_{2mp} contains a third moment which is closed by use of the stochastic process. Taking the limit in Equation 3.36, substituting the remaining means and variances and collecting like terms gives the stochastic parameterization for rain number concentration:

$$\begin{aligned} \frac{dq_r}{dt} = & \left(\frac{1}{2} + \mu_{2nf} + \mu_{2mf} + \mu_{2mp} \right) K_{cc} q_c N_c + K_{cr} q_c N_r + \\ & 2(\sigma_{2cnf}\xi_{2cnf} + \sigma_{2cmf}\xi_{2cmf} + \sigma_{2mp}\xi_{2mp}) K_{cc} q_c N_c \end{aligned} \quad (3.38)$$

This is the differential equation for rain aggregate mixing ratio, which contains the means of stochastic processes as physically meaningful parameters.

3.3 Parameter Reduction

Equations 3.16, 3.23, 3.31, and 3.38 form a system of stochastic differential equations which approximate the evolutions of the two moments of mass and number concentration of cloud and rain aggregates. They depend on a rather large number of parameters represented by the means and standard deviations of Ornstein Uhlenbeck processes. The parameter values can in principle be learned from data. As a first filtration, here we use detailed simulations based on the KCE (1) to infer the bulk behaviour of these parameters. This exercise will in particular allow us to eliminate the standard deviations from all the OU processes thus resulting in a set of deterministic equations in agreement with existing bulk cloud microphysics models (Khairoutdinov & Kogan 2000, Seifert & Beheng 2001, Franklin 2008). Numerical simulations were performed using Bott's Linear Flux Method (LFM) (Bott 1998). The

LFM is a popular and computationally efficient method of discretizing the kinetic collection equation. It simulates collisions of all possible two-droplet combinations at each time step, and evolves the mass density on an equidistant log-radius spectrum. Mass doubles every four bins (130 bins) and the radius of the smallest droplet represented on the spectrum we used is $0.589 \mu\text{m}$. The time step used is 1 second.

The detailed simulations are used in two independent ways to eliminate the fluctuation terms from the coupled set of ordinary differential equations shown in Equations 3.16, 3.23, 3.31, and 3.38. First, with respect to the representation of the stochastic processes as the sum of a mean and the product of a standard deviation and a homogeneous fluctuation, we show that the standard deviation term is at least one order of magnitude smaller than the mean term. This is shown using results from detailed simulations using the KCE with four different collision kernels. Thus we justify eliminating the fluctuation terms from Equations (3.16, 3.23, 3.31, 3.38). These fluctuation terms contain ξ . Second, when taking the mean of these four equations, second order fluctuation terms such as $\overline{\delta N_c \delta N_c}$ appear. We use the same detailed simulations to show that δN_c , δN_r , and δq_c , are at least three orders of magnitude smaller than $\overline{N_c}$, $\overline{N_r}$, and $\overline{q_c}$, respectively. The rain mixing ratio, q_r , does not appear on the right hand side of the stochastic bulk equations.

3.3.1 Temporal Mean of the Stochastic Parameters

In addition to identifying the setup used to show that $\delta N_c \ll \overline{N_c}$, $\delta N_r \ll \overline{N_r}$, and $\delta q_c \ll \overline{q_c}$ in Section 3.3, in this section we show that $\sigma_s \ll \mu_s$.

The kinetic collection equation (Eq. 2.1) is integrated for one hour and statistics are collected for the nine stochastic parameters contained in Equations 3.16, 3.23, 3.31, 3.38, defined as the sum of a mean and the product of a standard deviation and a homogeneous stochastic process: $X_s \equiv \mu_s + \sigma_s \xi_s$ where μ_s and σ_s are collected during the detailed simulations. The results are reported in Table 3.1, and with the exception of the hydrodynamic and turbulent A kernels for X_{4np} , the standard deviations are an order of magnitude smaller than the respective means. These exceptions are not troublesome because the rain self collection term can be analytically solved and written deterministically. The strength of the standard deviation term relative to the mean term is shown by the ratio σ/μ . The columns in Table 3.1 that are headed by this ratio show that the standard deviation term is at least

one order of magnitude smaller than the mean term. These simulations were done for four kernels: (i) the piece-wise polynomial kernel used by Seifert and Beheng (2001), (ii) a hydrodynamic kernel with collection efficiencies tabulated by Hall (1980), (iii) a turbulent kernel parameterized by Franklin (2007), and (iv) a turbulent kernel with tabulated values by Pinsky and Khain (2008). These four kernels (two turbulent, one hydrodynamic, and one analytic) were used to demonstrate the flexibility of applying a variety of different kernels in the new stochastic bulk parameterization.

Based on these results, the contribution of the random parts, containing ξ_x , of the processes X_s can be eliminated. This order of magnitude argument eliminates the terms containing the standard deviations, which are the very terms containing the stochastic fluctuations. The remaining four coupled equations are deterministic.

Table 3.1: Statistics of Fluctuations

Process	Polynomial		Hydrodynamic		Turbulent A		Turbulent B	
	μ	σ/μ	μ	σ/μ	μ	σ/μ	μ	σ/μ
X_{1nf}	-1.3e-01	-1.2e-04	-1.9e-01	-1.3e-01	-1.4e-01	-3.8e-02	-3.2e-01	-1.1e-01
X_{1np}	-1.5e-02	-7.7e-06	-8.2e-03	-3.5e-04	-1.2e-02	-5.2e-05	1.4e-03	6.9e-02
X_{2nf}	3.5e-02	8.5e-05	1.1e-02	4.9e-03	1.3e-02	1.6e-03	1.2e-02	7.0e-03
X_{2np}	1.5e-02	1.1e-05	8.0e-03	3.9e-04	1.2e-02	5.2e-05	3.4e-03	5.2e-03
X_{4np}	1.8e-01	2.3e-01	2.8e-01	3.4e-01	1.8e-01	3.1e-01	1.6e-01	4.5e-02
X_{1mf}	-3.8e-01	-3.8e-03	-4.8e-01	-2.1e-02	-3.6e-01	-5.7e-03	-7.1e-01	-1.4e-02
X_{1mp}	-7.7e-03	-2.8e-03	-7.3e-04	-4.9e-02	-5.2e-03	-1.3e-03	2.8e-02	5.2e-03
X_{2mf}	-3.0e-01	-8.5e-05	-3.4e-01	-6.7e-04	-3.3e-01	-4.2e-04	-2.9e-01	-1.2e-03
X_{2mp}	7.6e-03	2.9e-03	7.8e-04	4.3e-02	5.2e-03	1.3e-03	-6.8e-03	-2.6e-02

Table 3.1: The first column lists the nine stochastic parameters. The results from four kernels are given. The Polynomial kernel uses Long's sum of squared masses for cloud droplets and Golovin's linear sum of masses for rain droplets as presented in Siefert and Beheng (2010). The hydrodynamic kernel uses collision efficiencies given by Hall (1980). Turbulent A is Franklin's (2007) parameterized turbulent kernel, and Turbulent B is a tabulated kernel by Pinsky and Khain (2008).

3.3.2 Temporal Mean of the Evolving Quantities

As a confirmation of disregarding the fluctuation terms, we take the mean of these four stochastic equations (3.16, 3.23, 3.31, 3.38) and show that the fluctuation terms vanish. We substitute a mean and a fluctuation for the evolved quantities: $N_c = \overline{N_c} + \delta N_c$ where $\overline{N_c}$ is the ensemble mean. Taking the means of each evolved quantity (N_c , N_r , q_c , q_r) produces terms containing factors such as $\overline{\delta N_c \delta N_c}$ which are not necessarily zero. We use the detailed simulation results above to show that these terms are orders of magnitude smaller than the deterministic terms. Assuming ergodicity, $\overline{N_c}$, $\overline{N_r}$, $\overline{q_c}$, and $\overline{q_r}$ are computed by collecting N_c , N_r , q_c , and q_r at each time step. After the evolution is complete, the temporal average is calculated. The fluctuation terms are differences between the temporal average for each evolved quantity and the instantaneous value at each time step.

The graphical comparison of the fluctuations of the four evolving quantities to the respective temporal means of these quantities was done for four kernels: (a) the piece-wise polynomial kernel used by Seifert and Beheng (2001) shown in Figure 3.2a, (b) a hydrodynamic kernel with collection efficiencies tabulated by Hall (1980) shown in Figure 3.2b, (c) a turbulent kernel parameterized by Franklin (2007) shown in Figure 3.2c, and (d) a turbulent kernel with tabulated values by Pinsky and Khain (2008) shown in Figure 3.2d. These results are shown in the four graphs in Figure 3.2.

As shown in the four graphs in Figure 3.2, each of the evolving quantities, except for rain mixing ratio, has a maximum temporal fluctuation that is three orders of magnitude less than the quantities themselves. Consequently, we can alternatively take the approach of disregarding the cross terms containing, for example, $\overline{\delta N_c \delta N_c}$, thus providing a mean two-moment stochastic bulk rate parameterization shown in Equation 3.39. The four mean stochastic bulk rate equations contain nine parameters which are the means of (i) temporal fluctuations and (ii) temporal product fluctuations of number concentrations and mixing ratios in regions on the 2-D domain in Figure 3.1.

These spectral fluctuations arise from the approximation of number concentration density and of mixing ratio density as the sums of means and fluctuations according to Equations (3.3 and 3.4). These parameters are present in terms which represent collision processes: cloud self-collection,

Figure 3.2: Fluctuations from the Mean

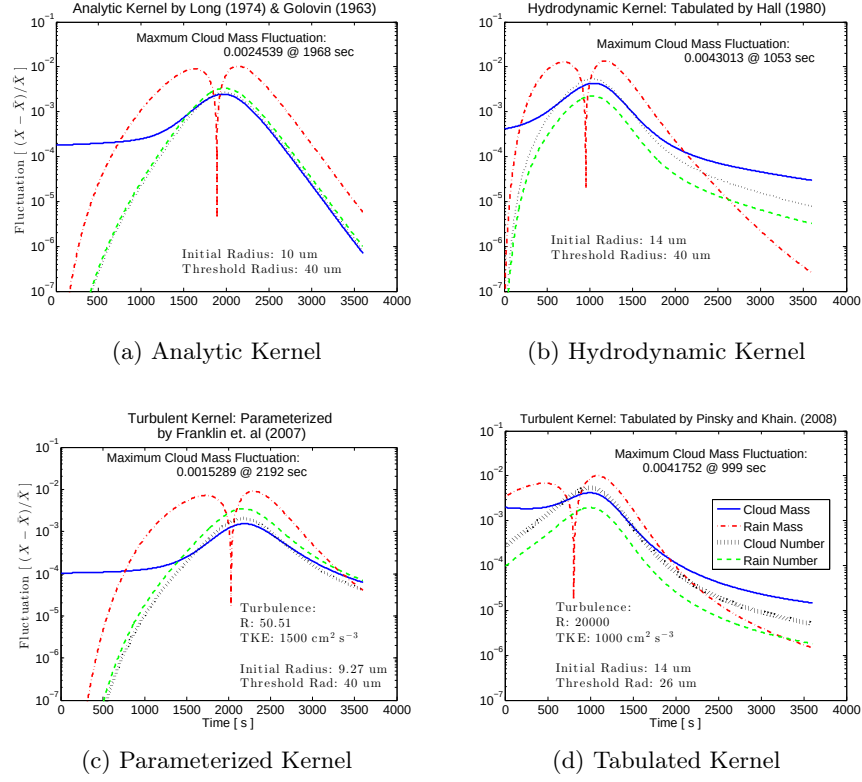


Figure 3.2: Ratio of the fluctuations to the mean value for each of the four evolved quantities: $\delta N_c / \overline{N_c}$, $\delta N_r / \overline{N_r}$, $\delta q_c / \overline{q_c}$, $\delta q_r / \overline{q_r}$ in each of the four plots. Assuming ergodicity, the value of each evolved quantity was averaged over a 60 minute simulation. The parameterization does not depend on the rain mixing ratio.

rain self-collection, auto conversion, and accretion.

$$\begin{aligned}
 \frac{d \overline{N_c}}{dt} &= - \left(\frac{3}{4} - \frac{2\mu_{1nf} + \mu_{1np}}{2} \right) K_{cc} \overline{N_c^2} - K_{cr} \overline{N_c N_r} \\
 \frac{d \overline{N_r}}{dt} &= \left(\frac{1}{4} + \frac{2\mu_{2nf} + \mu_{2np}}{2} \right) K_{cc} \overline{N_c^2} - \left(\frac{3}{4} - \mu_{4np} \right) K_{rr} \overline{N_r^2} \\
 \frac{d \overline{q_c}}{dt} &= - \left(\frac{1}{2} - (\mu_{1nf} + \mu_{1mf} + \mu_{1mp}) \right) K_{cc} \overline{q_c N_c} - K_{cr} \overline{q_c N_r} \\
 \frac{d \overline{q_r}}{dt} &= \left(\frac{1}{2} + \mu_{2nf} + \mu_{2mf} + \mu_{2mp} \right) K_{cc} \overline{q_c N_c} + K_{cr} \overline{q_c N_r}.
 \end{aligned} \tag{3.39}$$

The absence of q_r on the right hand side of Equation 3.39 shows that the aggregate number of rain droplets affects accretion, but the aggregate rain mixing ratio does not. This is in contrast to a model specifying individual droplet collisions whereby the mass of an individual rain droplet does affect the collection of a single cloud droplet (Pruppacher & Klett 1997).

3.3.3 Reduced Parameter Set

Each of the four differential equations in 3.39 contains linear combinations of statistics of the stochastic process. In four instances, the stochastic processes within a linear combination describe the statistics of a single collision and coalescence process. The means of fluctuations and of product fluctuations in a single region model the same phenomena and are linearly related in a single quantity. Since we are simply collecting values for these parameters from data and are not examining the effects of modifying one parameter relative to another parameter, combining parameters in this linear manner does not pose a problem.

$$\begin{aligned}
 \mu_{1n} &= 2\mu_{1nf} + \mu_{1np} && \text{fluctuation of number concentration in } \Omega_1 \\
 \mu_{2n} &= 2\mu_{2nf} + \mu_{2np} && \text{fluctuation of number concentration in } \Omega_2 \\
 \mu_{1m} &= \mu_{1nf} + \mu_{1mf} + \mu_{1mp} && \text{fluctuation of mixing in ratio in } \Omega_1 \\
 \mu_{2m} &= \mu_{2nf} + \mu_{2mf} + \mu_{2mp} && \text{fluctuation of mixing in ratio in } \Omega_2
 \end{aligned} \tag{3.40}$$

The stochastic bulk rate parameterization thus reduces to

$$\begin{aligned}
 \frac{dN_c}{dt} &= -\frac{1}{4}(3 - 2\mu_{1n})K_{cc}N_c^2 - K_{cr}N_cN_r \\
 \frac{dN_r}{dt} &= \frac{1}{4}(1 + 2\mu_{2n})K_{cc}N_c^2 - \left(\frac{3}{4} - \mu_{4np}\right)K_{rr}N_r^2 \\
 \frac{dq_c}{dt} &= -\left(\frac{1}{2} - \mu_{1m}\right)K_{cc}q_cN_c - K_{cr}q_cN_r \\
 \frac{dq_r}{dt} &= \left(\frac{1}{2} + \mu_{2m}\right)K_{cc}q_cN_c + K_{cr}q_cN_r.
 \end{aligned} \tag{3.41}$$

The bars in Equation (3.41) are dropped, but the meaning of the two-moment evolved quantities will remain unchanged.

3.3.4 Consistency of Number and Conservation of Mass

The first term on the right-hand-side of the cloud number (first) equation in (3.41) contains the effects of both self-collection and auto-conversion. Equation 3.14 distinguishes these effects, and separates the loss of droplets due to cloud self-collection from the gain of cloud droplets due to self-collection (i.e. the formation of a larger cloud droplet from two smaller cloud droplets). The first three terms on the right-hand-side of Equation 3.14 represent the increased number of cloud droplets due to self-collection. The fourth term ($-K_{cc}N_c^2$) represents the total loss (not net loss) of cloud droplets due to both processes. The coefficient of the first $K_{cc}N_c^2$ term in Equation 3.41 is rewritten to separate cloud droplet loss (cloud self-collection and auto conversion) from cloud droplet gain (only cloud self-collection): $-1/4(3 - 2\mu_{1n}) = [1/4 + (2/4)\mu_{1n}] - 1$.

Since two cloud droplets necessarily form either another cloud droplet or a new rain droplet, and we restrict collisions to binary ones, the following equation produces a constraint on the relationship between μ_{1n} and μ_{2n} . We equate the loss of cloud droplets to the gain of droplets resultant from that loss:

$$\underbrace{2 \left(\frac{1}{4} (1 + 2\mu_{1n}) \right)}_{\text{gain from cloud self-collection}} + \underbrace{2 \left(\frac{1}{4} (1 + 2\mu_{2n}) \right)}_{\text{gain from auto conversion}} - \underbrace{1}_{\text{loss}} = 0 \quad (3.42)$$

Equation (3.42) reduces to $\mu_{1n} = -\mu_{2n}$.

Conservation of mass is addressed by noting the similarity of the integration of the accretion terms (which span a complete aggregate), with the integration of the regions Ω_1 and Ω_2 when they are combined. The resultant integral also spans a complete aggregate and can be solved analytically. Particularly, we have $n(z)n(y)K_{cc}$ integrated over the combined region yielding $K_{cc}N_c^2$. The first loss term from Equation 3.12 integrates to $-K_{cc}N_c^2$. Combining the mass gain and mass loss terms we have:

$$K_{cc}N_c^2 - K_{cc}N_c^2 = 0. \quad (3.43)$$

Separating the integrals for mixing ratio over domains Ω_1 and Ω_2 , the two resultant integrals do not span a complete aggregate and we must employ stochastic processes to close the equations as was detailed in Sections (3.2.3 and 3.2.4). One stochastic term, μ_{1m} , arises from the linear combination of the second, third, and fourth terms on the right hand side of Equation 3.29.

The other stochastic term, μ_{2m} , arises from the linear combination of the second, third, and fourth terms on the right hand side of Equation 3.36. The deterministic term is the fifth term on the right hand side of Equation 3.29. The coefficients of these three mixing ratio terms, which contain $K_{cc}N_c^2$, are set equal to the right hand side of Equation 3.43:

$$\left(\frac{1}{2} + \mu_{1m}\right) + \left(\frac{1}{2} + \mu_{2m}\right) - 1 = 0.$$

From which follows: $\mu_{1m} = -\mu_{2m}$. Rewriting the four stochastic bulk rate equations using three independent parameters yields

$$\begin{aligned} \frac{dN_c}{dt} &= -\frac{1}{4}(3 - 2\mu_{1n})K_{cc}N_c^2 - K_{cr}N_cN_r \\ \frac{dN_r}{dt} &= \frac{1}{4}(1 - 2\mu_{1n})K_{cc}N_c^2 - \left(\frac{3}{4} - \mu_{4np}\right)K_{rr}N_r^2 \\ \frac{dq_c}{dt} &= -\left(\frac{1}{2} - \mu_{1m}\right)K_{cc}q_cN_c - K_{cr}q_cN_r \\ \frac{dq_r}{dt} &= \left(\frac{1}{2} - \mu_{1m}\right)K_{cc}q_cN_c + K_{cr}q_cN_r \end{aligned} \tag{3.44}$$

Conservation of mass for auto conversion follows intrinsically from the derivations and is clearly shown by the similarity of the first terms on the right hand sides of the mixing ratio equations in Equation 3.44. Conservation of mass for accretion follows from the $K_{cr}q_cN_r$ terms in these same two equations.

The physical meanings and effects of these final three parameters are as follows:

μ_{1n}	controls the strength of cloud self-collection relative to auto-conversion
μ_{1m}	controls the strength of auto-conversion
μ_{4np}	controls the strength of rain self-collection.

Bounds on the values of these stochastic parameters will be established in Chapter 4.

3.4 Summary

Here, we presented a stochastic bulk parameterization of collision and coalescence which does not rely on any probability distribution of the droplet size spectrum, but rather assumes that there is a distribution and it has a mean. All of the parameters in the stochastic parameterization have physical meaning, and their values can be recovered from data. The parameters represent uncertainties in the first and second moments of time series of aggregate fluctuations of mixing ratio and number over defined portions of the droplet size spectrum.

The flexibility of using any collision kernel in the stochastic parameterization comes at the expense of retaining only the zeroth order term in the 2D Taylor expansion of the collision kernel centred at either the cloud, or rain, mean mass. This flexibility requires that the value of the collision kernel be computed, or retrieved from a look-up table, while the stochastic parameterization is being utilized by a climate model. The stochastic bulk parameterization of cloud microphysical processes contain only three stochastic parameters, one for each three collision processes: cloud-self collection, rain self-collection, and auto conversion.

The truncation of the collision kernel and the novel representation of the distribution of the droplet size spectrum permitted the uncertainties of the instantaneous droplet size densities to be modelled as points in a stochastic process. Thus providing (to the author's knowledge) an unprecedented method to solve the closure problem in the integro-differential equation known as the Kinetic Collection Equation, presented in Chapter 2 as Equation 2.1.

4 | Validation

In Chapter 3, we systematically decomposed the number concentration density and mixing ratio density into state-space means and sets of point-wise fluctuations within cloud and rain droplet aggregates. We then summed the fluctuations over defined intervals and considered each instantaneous sum to be a point in a stochastic process. Closure of the kinetic collection equation was achieved by representing higher moments as functions of the point-wise fluctuations. The stochastic processes were defined as a mean and a standard deviation, and order of magnitude arguments eliminated the standard deviation terms. The means of the stochastic processes are parameters. Consistency of number and conservation of mass were used to reduce the degrees of freedom of the stochastic parameterization.

4.1 Auto conversion Parameters

Number concentration and mixing ratio are related by Equation 2.16 ($\bar{x} = q/N$). Writing the four stochastic bulk rate equations using three independent parameters yields,

$$\begin{aligned}
 \frac{dN_c}{dt} &= -\frac{1}{4}(3 - 2\mu_{1n})K_{cc}N_c^2 - K_{cr}N_cN_r \\
 \frac{dN_r}{dt} &= \frac{1}{4}(1 - 2\mu_{1n})K_{cc}N_c^2 - \left(\frac{3}{4} - \mu_{4np}\right)K_{rr}N_r^2 \\
 \frac{dq_c}{dt} &= -\left(\frac{1}{2} - \mu_{1m}\right)K_{cc}q_cN_c - K_{cr}q_cN_r \\
 \frac{dq_r}{dt} &= \left(\frac{1}{2} - \mu_{1m}\right)K_{cc}q_cN_c + K_{cr}q_cN_r.
 \end{aligned} \tag{4.1}$$

The physical meanings and effects of these parameters are as follows.

μ_{1n}	controls the strength of cloud self-collection relative to auto-conversion,
μ_{1m}	controls the strength of auto-conversion,
μ_{4np}	controls the strength of rain self-collection.

The derivations and details which resulted in the stochastic parameterization presented in Equation 4.1 are given in the previous chapter. The values of each of the three stochastic parameters could be acquired from the results of a bin-pair interaction detailed method.

The ‘SDE-based’ stochastic model is general enough for any kernel, but here we validate it by applying the piece-wise polynomial kernel given in Equation 2.3, and graphically acquire values of the stochastic parameters after constraining the stochastic parameters using conservation of mass, consistency of number, and the definition of auto conversion. We compare it to Siefert and Beheng’s model (2001) using results from Bott’s detailed method as a benchmark. The values for results driven by a hydrodynamic kernel and by two turbulent kernels are presented in Chapter 5.

4.1.1 Physical Limits: Consistency of Number and Conservation of Mass

Since the kernel (K_{cc}) and number concentration (N_c) are non-negative, they are omitted from the following expressions. Cloud self-collection and auto-conversion each decrease cloud number concentration, thus: $\left(\frac{3}{4} - \frac{1}{2}\mu_{1n}\right) \geq 0$. Auto-conversion will cause rain number concentration to increase, thus: $\frac{1}{4}(1 - 2\mu_{1n}) \geq 0$. Both of these conditions are satisfied by $\mu_{1n} \leq 1/2$. Because cloud self-collection does not affect rain number concentration, the cloud loss will be at least twice the rain increase:

$$\left(\frac{3}{4} - \frac{1}{2}\mu_{1n}\right) \geq 2\left(\frac{1}{4}(1 - 2\mu_{1n})\right). \quad (4.2)$$

This results in $-1/2 \leq \mu_{1n}$. Auto conversion will decrease cloud mixing ratio and cause rain mixing ratio to increase, thus: $\left(\frac{1}{2} - \mu_{1m}\right) \geq 0$. Thus

we have

$$-\frac{1}{2} \leq \mu_{1n} \leq \frac{1}{2} \quad \text{and} \quad \mu_{1m} \leq \frac{1}{2}. \quad (4.3)$$

The bounds on the two parameters related to cloud self-collection and auto conversion due to accuracy of number and conservation of mass can be represented on a single graph shown in the picture on the left in Figure 4.1. The lower bound on μ_{1m} , or the strength of auto-conversion, is dependent on the distance of the mean cloud radius to the threshold radius: the closer they are together the stronger the auto conversion rate and a smaller mean cloud radius demands a weaker auto-conversion rate. This specific relationship can be quantified by examining the coefficients of the K_{cc} terms in Equation 4.1.

Additionally, rain self-collection decreases rain number concentration and is constrained by the following inequality:

$$\mu_{4nc} \leq \frac{3}{4}.$$

4.1.2 Autoconversion and the Threshold Droplet Size

There is a need to constrain the auto conversion parameter (Wood 2005). Franklin (2007) and Khairoutdinov and Kogan (2000) have constrained it in an ad-hoc manner using best fit methods to DNS and to a detailed, bin based method, respectively. Neither their constraints nor their parameters have physically based meanings (Franklin, Vaillancourt & Yau 2007, Khairoutdinov & Kogan 2000). In the stochastic bulk parameterization, the auto conversion parameter is further constrained using the relationship between the mean cloud radius and the threshold radius, and the definition of auto-conversion; namely, that pre-collision, and post-collision, droplets must be smaller than and larger than the threshold radius, respectively. All of the mass in an auto conversion collision moves from cloud to rain.

The effect that auto-conversion has on the rain spectrum is described by the first terms on the right hand side of the second and fourth equations in Equation 4.1. These are related to number and mixing ratio, respectively. Therefore, given $\frac{q_c}{N_c} = \bar{x}_c$, the equation requiring that post-collision droplets have a mass greater than the mass of the threshold droplet is

$$x^* \leq \frac{(\frac{1}{2} - \mu_{1m})K_{cc}q_cN_c}{\frac{1}{4}(1 - 2\mu_{1n})K_{cc}N_c^2} = \frac{(\frac{1}{2} - \mu_{1m})}{\frac{1}{4}(1 - 2\mu_{1n})}\bar{x}_c.$$

Figure 4.1: Constraining the Auto Conversion Parameter in the μ_{1n} - μ_{1m} plane

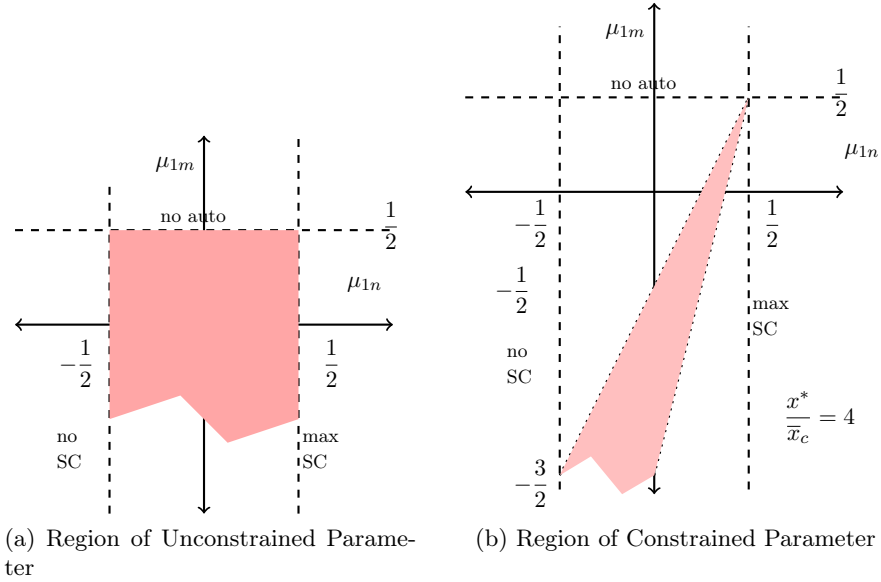


Figure 4.1: Physical constraints (conservation of mass and accuracy of number) on the stochastic parameters associated with cloud self-collection and with auto-conversion. The triangle in (b) extends to $(-\frac{1}{2}, -\frac{7}{2})$, and is representative of the situation when the threshold radius is $40 \mu\text{m}$ and the mean cloud radius is $25.2 \mu\text{m}$. The mean cloud radius $\bar{r}_c$ and mean cloud mass $\bar{x}_c$ are related by $\bar{x}_c = 4/3\pi\rho\bar{r}_c^3$, and similarly for the threshold mass and radius.

The region in Figure 4.1a shows valid values of stochastic parameters before applying the relationship of auto conversion to the threshold droplet mass. Letting $\beta = \frac{x^*}{\bar{x}_c}$ and solving for μ_{1m} gives the equation for the upper boundary of the triangular region in Figure 4.1b:

$$\mu_{1m} \leq \frac{2 - \beta}{4} + \frac{\beta}{2}\mu_{1n}. \quad (4.4)$$

The lower boundary is constructed from the requirement that pre-collision droplets are each smaller than the threshold droplet. This is accomplished algebraically by considering that the number of cloud droplets lost to auto-conversion is twice the number rain droplets produced by the same process. The number of cloud droplets lost is given by doubling the first term on the rhs if the second differential equation in Equation 4.1 and proceeding similarly as for the upper boundary, we have

$$x^* \geq \frac{(\frac{1}{2} - \mu_{1m})K_{cc}q_cN_c}{\frac{1}{2}(1 - 2\mu_{1n})K_{cc}N_c^2} = \frac{(\frac{1}{2} - \mu_{1m})}{\frac{1}{2}(1 - 2\mu_{1n})}\bar{x}_c,$$

which yields the lower bound on the triangular region in the graph on the right in Figure 4.1:

$$\mu_{1m} \geq \frac{1 - \beta}{2} + \beta\mu_{1n}. \quad (4.5)$$

Using $\beta = 4$ as is depicted in Figure 4.1b, we get for the lower and upper bounds, respectively:

$$\mu_{1m} \geq -\frac{3}{2} + 4\mu_{1n} \quad \text{and} \quad \mu_{1m} \leq -\frac{1}{2} + 2\mu_{1n}.$$

When a cloud is young or heavily polluted, we can expect β to be large and the values for the stochastic parameters to be near the upper right corner of the region in the $\mu_{1n} - \mu_{1m}$ plane. This demands strong cloud self-collection and weak auto-conversion, which is to be expected when the cloud droplets are primarily small. By contrast in a mature cloud or one with primarily large droplets, β will be smaller and the region giving values for μ_{1n} & μ_{1m} will be different.

The strength of cloud self-collection is determined from the relationship between the first terms in the first two differential equations in Equation 4.1. When the value of μ_{1n} is chosen such that the first term in the equation for N_r is zero (i.e. $\mu_{1n} = 1/2$), none of the droplets subtracted in the equation for N_c creates rain droplets and thus all of them create cloud droplets and cloud

self collection is maximized. Consequently, auto-conversion is non-existent and the region of valid values for μ_{1n} & μ_{1m} collapses to intersection of the lines for max cloud self-collection and no auto-conversion (i.e. $\mu_{1n} = 1/2$ and $\mu_{1m} = 1/2$).

Alternately, when the value of μ_{1n} is chosen such that all of the droplets subtracted in the evolution of N_c in Equation 4.1 produce rain droplets in the second equation, N_r (i.e. $\mu_{1n} = -1/2$), cloud self-collection is non-existent and auto conversion is maximized. In other words, the coefficient of the first term on the right hand side of the dN_c/dt equation in Equation 4.1 is twice the coefficient of the first term on the right hand side of the dN_r/dt equation because two cloud droplets produce one rain droplet in the process of auto-conversion.

4.1.3 Constraint on the Autoconversion Parameter

The constraining of the auto conversion parameter is possible because the four auto conversion terms are derived independently. The independent derivations allow the ratios of mixing ratio and number concentration to be compared to the threshold mass. This is not possible when one auto conversion term is a function of another auto conversion term. The equations for the two sloped sides of the triangle in Figure 4.1 appear because of the independent derivations of the number concentration and mixing ratio equations. The third side is resultant from the two cloud droplet terms for number being derived independently.

Conservation of mass and consistency of number create the same bounds as the upper right vertex of the triangle in Figure 4.1 which is the location on the graph representative of strong cloud self-collection and weak (to non-existent) auto conversion. Stochastic parameter values in this area of the triangle model a polluted cloud with many small droplets. Consistency of number constrains the maximum and minimum value of the cloud self-collection parameter. Conservation of mass only constrains the maximal value (weakest strength) of the auto conversion parameter.

The maximum strength of auto conversion is limited by the lower vertex of the triangle. Auto conversion is maximal when the mean cloud radius is close to the threshold radius which gives minimal values to μ_{1m} in the two vertices on the left side of the triangle which corroborates some of Franklin's results (Franklin 2008). Alternatively, when cloud self-collection is strong,

the lower bounds on the value of the auto conversion parameter is restricted further than the apparent bounds given by the perimeter of the triangle: a strong cloud self-collection parameter ($\mu_{1n} \approx 1/2 - \epsilon$) disallows μ_{1m} taking values from the lower area of the triangle. Thus μ_{1m} is constrained to be ‘near’ 0.5, i.e. modelling a weaker auto conversion rate.

4.2 Model with Piece-Wise Kernel

Siefert and Beheng developed in 2001 a parameterization that models the effects of collision and coalescence on the evolution of four bulk cloud micro-physical properties. Their parameterization was subsequently used in 2010 in a 1-D rain shaft model that included droplet breakup and condensation and evaporation (Seifert & Stevens 2010).

The terms in Seifert and Beheng’s model which is shown in Equation 2.20 are labeled by the collision processes that they model. Auto conversion occurs in each of the four evolving quantities and accretion occurs in three equations. The coefficients of seven of the eight terms are dependent on an ad-hoc shape parameter ν , and all are dependent on one or more of six other parameters ($k_c, k_r, \tau, x^*, \bar{x}_c, \bar{x}_r$).

Although Siefert and Beheng omitted $\frac{\nu + 2 \bar{x}_c}{\nu + 1 \bar{x}_r}$ from the accretion terms in their final parameterization, this factor is included in Γ_{acct} in Equation 2.18 for completeness, and an equivalent term appears in precisely the same location in the stochastic model once the same piece-wise polynomial kernel is applied. These eight coefficients, excluding the collision kernels, are equated with the coefficients of the corresponding terms in the stochastic parameterization.

4.2.1 Reproduce Existing Parameterization

We reproduce a bulk model driven by a piece-wise kernel. Applying a different kernel, say a hydrodynamic or a turbulent one, would yield a different parameterization. The piece-wise polynomial kernel shown in Equation 2.3 is applied to the stochastic bulk parameterization Equation 4.1. This analytic kernel introduces the centroids of the cloud and rain aggregates into the stochastic parameterization which appear in the accretion terms. The

stochastic bulk parameterization of cloud microphysical collision processes driven by the Long-Golovin kernel is:

$$\begin{aligned}
\frac{dN_c}{dt} &= -\frac{1}{2}(3 - 2\mu_{1n})k_c q_c^2 - \left(1 + \frac{\bar{x}_c}{\bar{x}_r}\right)k_r q_r N_c \\
\frac{dN_r}{dt} &= \frac{1}{2}(1 - 2\mu_{1n})k_c q_c^2 - \left(\frac{3}{2} - 2\mu_{4np}\right)k_r q_r N_r \\
\frac{dq_c}{dt} &= -(1 - 2\mu_{1m})\bar{x}_c k_c q_c^2 - \left(1 + \frac{\bar{x}_c}{\bar{x}_r}\right)k_r q_c q_r \\
\frac{dq_r}{dt} &= (1 - 2\mu_{1m})\bar{x}_c k_c q_c^2 + \left(1 + \frac{\bar{x}_c}{\bar{x}_r}\right)k_r q_c q_r.
\end{aligned} \tag{4.6}$$

A term-by-term comparison of Equations (2.20 and 4.6) reveals an exact match regarding the dependencies on aggregate number concentrations (N_c and N_r) and mixing ratios (q_c and q_r). The coefficients of corresponding terms are equated to determine the range of the stochastic parameters as functions of the range of values that Siefert and Beheng gave to their parameters, τ and ν .

4.2.2 Select Stochastic Parameter Values

We make a systematic comparison by matching corresponding terms. In both Seifert and Beheng's parameterization and the stochastic parameterization, the auto conversion terms for cloud and rain mixing ratio are equal with the opposite sign. The same is true for the cloud and rain accretion terms. Consequently, with regard to mixing ratio, only the auto conversion and accretion terms that evolve rain mixing ratio are compared because the cloud terms have been restricted by conservation of mass. The stochastic term for rain self-collection is compared with the corresponding term in the Seifert and Beheng parameterization. The subsequent calculations use a threshold radius of $40\mu\text{m}$ and a mean cloud radius of $10\mu\text{m}$. The auto conversion term for rain number concentration contains a stochastic parameter (μ_{1n}) that is a different parameter than the one for mixing ratio (μ_{1m}), and so the rain number coefficients are compared. The first terms in both cloud number concentration equations combine the processes of auto conversion and cloud self-collection. These are compared; however, they contain the same stochastic parameter (μ_{1n}) as in rain number concentration. The more meaningful range of values for μ_{1n} is derived from cloud number concentration. The physically permissible values of the stochastic parameters that

reproduce Seifert and Beheng's bulk parameterization are collected tabularly and displayed graphically in Section 4.2.3.

Auto conversion: mixing ratio

The coefficients for auto conversion of rain (and cloud) mixing ratio are set equal,

$$(1 - 2\mu_{1m}) \bar{x}_c = \frac{1}{20} \frac{(\bar{x}_c)^2}{x^*} \frac{(\nu + 2)(\nu + 4)}{(\nu + 1)^2} \left[1 + \frac{\Phi_{\text{au}}(\tau)}{(1 - \tau)^2} \right],$$

and solved for the stochastic parameter:

$$\mu_{1m} = \frac{1}{2} - \frac{1}{40} \frac{\bar{x}_c}{x^*} \frac{(\nu + 2)(\nu + 4)}{(\nu + 1)^2} \left[1 + \frac{\Phi_{\text{au}}(\tau)}{(1 - \tau)^2} \right].$$

The shape parameter ν takes values over the interval $(0, 3)$. The function, $\Phi_{\text{au}}(\tau)$, of the time parameter, $\tau = q_r/(q_c + q_r)$, has a maximum at $\Phi_{\text{au}}(\tau) \approx 0.27$. The particular form of $\Phi_{\text{au}}(\tau)$ is not relevant to this research and is therefore omitted. The bounds that Seifert and Beheng set for ν and τ and the maximum of $\Phi_{\text{au}}(\tau)$ are used to determine the range of the stochastic parameter governing the strength of auto conversion:

$$0.20094 \leq \mu_{1m} \leq 0.49829. \quad (4.7)$$

This range is displayed graphically (blue patterned region) in Figure 4.2.

Auto conversion: rain number concentration

Equating the corresponding auto-conversion terms for rain number concentration gives

$$\frac{1}{2} (1 - 2\mu_{1n}) = \frac{1}{20} \left(\frac{\bar{x}_c}{x^*} \right)^2 \frac{(\nu + 2)(\nu + 4)}{(\nu + 1)^2} \left[1 + \frac{\Phi_{\text{au}}(\tau)}{(1 - \tau)^2} \right].$$

Solving for the stochastic parameter gives:

$$\mu_{1n} = \frac{1}{2} - \frac{1}{20} \left(\frac{\bar{x}_c}{x^*} \right)^2 \frac{(\nu + 2)(\nu + 4)}{(\nu + 1)^2} \left[1 + \frac{\Phi_{\text{au}}(\tau)}{(1 - \tau)^2} \right] \quad (4.8)$$

The stochastic parameter governing the strength of cloud self-collection relative to auto conversion is constrained by Equation 4.8 to values less than but close to the maximum permissible value: $0.49066 \leq \mu_{1n} \leq 0.49997$. The lower bound is consistent with the value of μ_{1n} when $\mu_{1m} = 0.20094$ given the constraint that a rain droplet produced during an auto conversion process must be larger than the threshold mass x^* .

Auto conversion: cloud number concentration

Equating the corresponding auto-conversion terms for cloud number concentration gives:

$$\frac{1}{2}(3 - 2\mu_{1n}) = \frac{\nu + 2}{\nu + 1} \quad \text{which implies} \quad \mu_{1n} = \frac{3}{2} - \frac{\nu + 2}{\nu + 1}$$

For any value of ν specified by Siefert and Beheng ($0 \leq \nu \leq 3$), the bounds on the stochastic parameter is readily calculated: $0.25 \leq \mu_{1n} \leq 0.5$. The derivation of their auto conversion term for rain number concentration uses the assumptions that $\bar{x}_r \approx x^*$ and $\bar{x}_c \ll \bar{x}^*$. These assumptions may be true during the early stages of a cloud's lifetime, but are less likely to be true in a mature cloud. Therefore, the lower bound ($\mu_{1n} = 0.25$) derived in this section will be used to constrain this stochastic parameter. Since the upper bound based on cloud number concentration terminates auto conversion, the upper bound ($\mu_{1n} = 0.49997$) based on rain number concentration will be used.

$$0.25 \leq \mu_{1n} \leq 0.49997 \quad (4.9)$$

This range is displayed graphically (blue patterned region) in Figure 4.2.

Accretion

None of the three accretion terms in the stochastic bulk parameterization contain stochastic parameters. Except for the sign, the coefficients for the three accretion terms are identical and we have

$$\left(1 + \frac{\bar{x}_c}{\bar{x}_r}\right) = \Phi_{ac}(\tau) \left(1 + \frac{\nu + 2}{\nu + 1} \frac{\bar{x}_c}{\bar{x}_r}\right).$$

The non-dimensional time function, $\Phi_{ac}(\tau)$, is an ad-hoc parameter that was included after completing their derivations, and the shape parameter, ν , is a by-product of the assumed droplet gamma distribution. The particular form of $\Phi_{ac}(\tau)$ is not relevant to this research and is therefore omitted. Seifert and Beheng removed the shape parameter function from the accretion terms in their final parameterization scheme. There are no arbitrary or ad-hoc parameters in the stochastic bulk rate parameterization. Consequently, the only difference between the stochastic parameterization and their 'adjusted' parameterization is the non-dimensional time function which was added after derivations were complete. This function serves to slow accretion during the early and late stages of a cloud's lifetime and accelerate accretion during the middle stages, maximally when $\frac{q_r}{q_c + q_r} \approx 0.27$.

A first order truncation of the collision kernel in the stochastic parameterization may give stochastic parameters in the accretion terms which may serve the same purpose as their non-dimensional time function. However, all stochastic parameters are associated with concrete physical meanings. Trading the higher order Taylor expansion terms of the collision kernel for the simplicity of the ‘SDE-based’ stochastically derived set of coupled ODE’s and omitting the ad-hoc non-dimensional parameter inclusions, all accretion terms are identical.

Rain self-collection: number concentration

Equating the corresponding rain self-collection terms for number concentration gives:

$$\frac{3}{2} - 2\mu_{4nc} = 1, \quad \text{thus} \quad \mu_{4nc} = \frac{1}{4} < \frac{3}{4} \quad \text{as required.} \quad (4.10)$$

Table 4.1: Stochastic Parameter Bounds: Polluted Cloud

Parameter	Lower	Condition	Upper
μ_{1n}	0.49066	—	0.49997
μ_{1m}	0.20094	$0.49066 \leq \mu_{1n} \leq 0.49533$	$32\mu_{1n}-15.5$
	$64\mu_{1n}-31.5$	$0.49533 \leq \mu_{1n} \leq 0.49995$	
		$0.49995 \leq \mu_{1n} \leq 0.49997$	
μ_{4np}		=0.25	0.49829

Table 4.1: The bounds of the auto conversion μ_{1m} and cloud self-collection μ_{1n} parameters for simulations with a threshold radius of $40\mu\text{m}$ and a mean cloud mass of $10\mu\text{m}$.

4.2.3 Regions of Validity in Parameter Space

The stochastic parameterization reproduces Siefert and Beheng’s parameterization when μ_{1n} , μ_{1m} , and μ_{4np} are restricted to values identified in Equations (4.7, 4.9, and 4.10). These restrictions are formed by making the stochastic parameters to be functions of Siefert and Beheng’s parameters: ν and τ . Equations (4.7 and 4.9) bound the values of μ_{1n} and μ_{1m} and create a 2D space of parameter values which is displayed as the patterned (blue) in

Figure 4.2: Region of Validity in μ_{1n} - μ_{1m} : Polluted Cloud

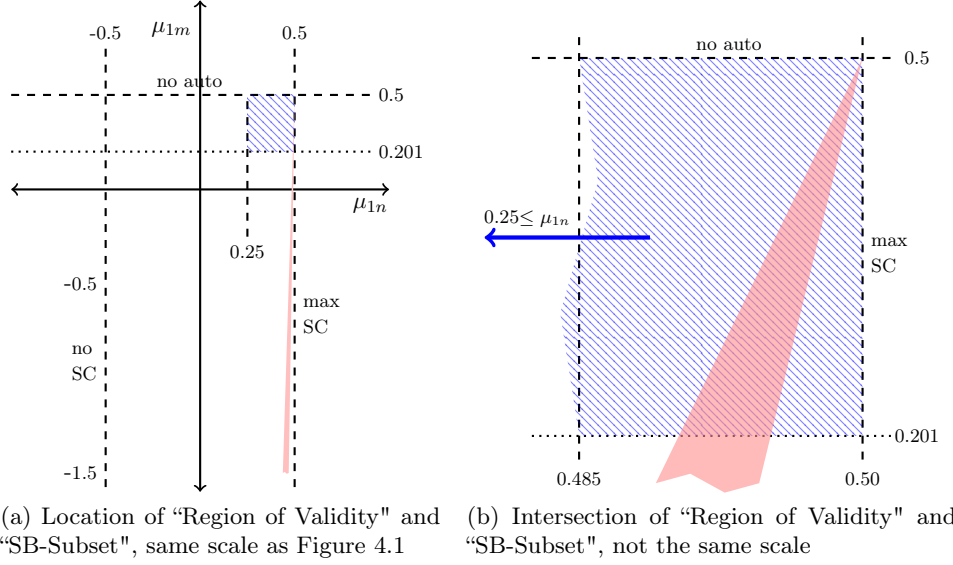


Figure 4.2: The graph (a) is similar to and has the same scale as the two graphs in Figure 4.1. The lower bound of the triangle in the figures extends to $(-1/2, -63.5)$, and is representative of the situation when the threshold radius is $40\mu\text{m}$ and the mean cloud radius is $10\mu\text{m}$. The graph (b) is a blow-up of the intersection of (i) the region (solid, red) of physically permissible values of stochastic parameters and (ii) the region (patterned, blue) of values that reproduce Seifert and Beheng's parameterization for their range of parameters. The orientation of the intersection is given by $x^*/\bar{x}_c = 64$.

both graphs in Figure 4.2. Values within this region control the strength of cloud self-collection and auto conversion. This rectangular region intersects the triangular (solid, red) region formed using the physical constraints from Sections 4.1.1 and 4.1.2.

This intersection is a pentagonal region with vertices (counter-clockwise from lower left in the graph on the right of Figure 4.2 at $(0.49066, 0.20094)$, $(0.49533, 0.20094)$, $(0.49997, 0.49808)$, $(0.49997, 0.49829)$, and $(0.49995, 0.49829)$). This pentagon is most clearly depicted in Figure 4.4 when discussing sensitivity in Section 4.3.3. The bounds of values of μ_{1n} and μ_{1m} in this pentagonal region are listed in Table 4.1. Three of the sides of the pentagon are apparent in Figure 4.2b. The graphs in Figure 4.2 and the values in Table 4.1 are derived using a threshold radius of $40\mu\text{m}$ and an initial mean cloud radius of $10\mu\text{m}$ which is representative of a terrestrial or

Figure 4.3: Region of Validity in μ_{1n} - μ_{1m} : Clean Cloud

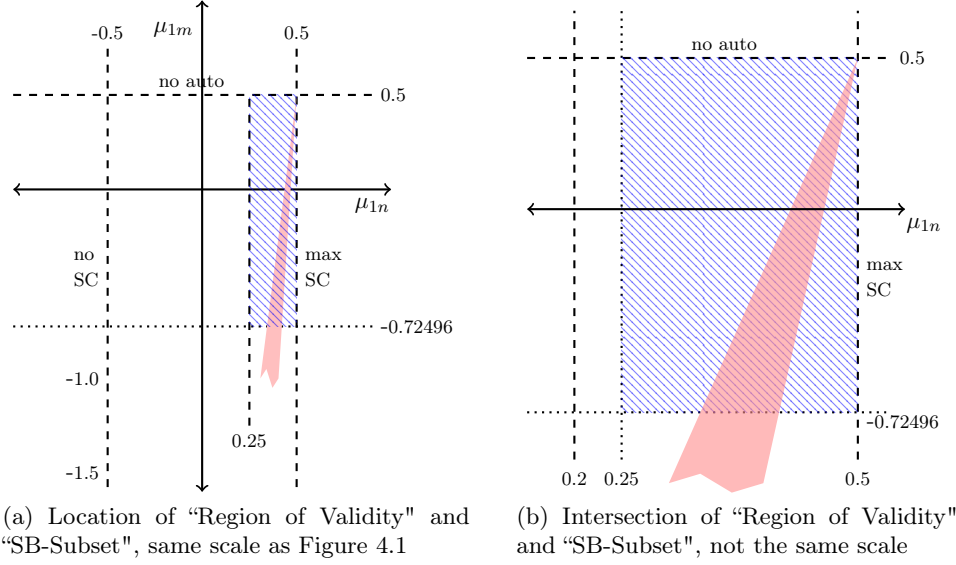


Figure 4.3: Same as Figure 4.2 except the lower bound of the triangle in the figures extends to $(-1/2, -15.125)$, and is representative of the situation when the threshold radius is $40\mu\text{m}$ and the mean cloud radius is $16\mu\text{m}$. The orientation of the intersection is given by $x^*/\bar{x}_c = 15.625$.

‘polluted’ cloud. The equations for the two sloped sides of the solid, red triangle in Figure 4.2 are given as upper and lower bounds of μ_{1m} in Table 4.1.

For the ‘clean’ cloud, an initial mean cloud radius of $16\mu\text{m}$ is used with the same threshold radius. The bounds of the stochastic parameters for a ‘clean’ or marine cloud when the evolution is driven by the Long-Golovin kernel is shown graphically in Figure 4.3 and numerically in Table 4.2. In a ‘clean’ cloud, the ratio $\frac{\bar{x}_c}{x^*}$ forces a sufficiently smaller upper bound on μ_{1n} (weaker cloud self-collection and stronger auto conversion) such that the intersecting region is a quadrilateral. The vertices (counter-clockwise from lower left in Figure 4.3b of this four-sided figure are $(0.34320, -0.72496)$, $(0.4216, -0.72496)$, $(0.49300, 0.39063)$, and $(0.49300, 0.44531)$. Three of the sides of the quadrilateral are apparent in Figure 4.3. This quadrilateral is most clearly depicted in Figure 4.5 when discussing sensitivity in Section 4.3.3. The equations for the two sloped sides of the solid, red triangle are given as upper and lower bounds of μ_{1m} in Table 4.2.

Table 4.2: Stochastic Parameter Bounds: Clean Cloud

Parameter	Lower	Condition	Upper
μ_{1n}	0.34320	—	0.49300
μ_{1m}	-0.7250	$0.34320 \leq \mu_{1n} \leq 0.42160$	$7.8125\mu_{1n}-3.40625$
	$15.625\mu_{1n}-7.3125$	$0.42160 \leq \mu_{1n} \leq 0.49300$	$7.8125\mu_{1n}-3.40625$
μ_{4np}		$=0.25$	

Table 4.2: The same as Table 4.1 except with a mean cloud mass of $16\mu\text{m}$.

4.3 Numerical Simulations

The intersection of the rectangular (blue), ‘SB subset region,’ and triangular (red), ‘region of validity’ of parameter values form polygons. Parameter values within these polygons both represent physically real conditions and are a function of Seifert and Beheng’s parameters. We examine the evolution of aggregate number concentration and mass by the stochastic bulk model by (i) varying the cloud-self collection and auto conversion parameters within the intersection of the ‘region of validity’ and the ‘SB subset region,’ (ii) observing the time required to convert 50% of cloud droplet mass to rain droplets within this intersection, and (iii) comparing the “50% time” to results from detailed simulations of the KCE equation using the Linear Flux Method (Bott 1998). We show the effects of macroscopic cloud properties within the red-blue intersection of by calculating the rain droplet radius at the first time step and the radius at the time of 50% conversion. Finally, sensitivity tests are performed with respect to the time step and the rain self-collection parameter in Section 4.3.3.

4.3.1 Model Setup

We perform simulations for two cloud types: polluted and clean. The polluted clouds have an initial mean cloud droplet radius of $10\mu\text{m}$ which is 239 droplets per cm^3 given the typical liquid water content of 1 g cm^{-3} . Clean clouds have an initial mean cloud droplet radius of $16\mu\text{m}$ and a correspond-

ing droplet concentration of 58.3 cm^{-3} . The third stochastic parameter μ_{4np} , regulating rain self-collection is fixed at 0.25. All simulations use 3rd order Adams-Bashforth with Improved Euler and 2nd order Adams-Bashforth for the first and second time step, respectively.

For the detailed benchmark simulations, we use the Linear Flux Method by Bott (1998) with 70 bins that double every second bin. The smallest droplet radius on the discretized spectrum is $1 \text{ }\mu\text{m}$ and the largest is 3.251 mm .

4.3.2 Results

Three types of results are shown for each cloud type. We present contour graphs of the 50% conversion time that span relevant portions of the intersection of the the region of validity and the ‘SB subset region.’ The relevant portions are where changes in the 50% conversion time as a function of the cloud self-collection and auto conversion parameters are noticeable. This occurs where cloud self-collection is stronger and auto conversion is weaker, where μ_{1m} and μ_{1n} are nearby and slightly less than 0.50. Within this same region in the (μ_{1m}, μ_{1n}) space we present contour graphs showing how the mean rain radius, at the 50% conversion time, changes as the stochastic parameters are varied. Additionally we present another type of contour graph that shows how the mean rain radius at the first time step varies as a function of (μ_{1m}, μ_{1n}) . The third result is a set of curves of mass-over-time graphs comparing the conversion of cloud to rain mass by the stochastic model, Seifert and Beheng’s model, and the detailed Linear Flux Method (Bott 1998).

50% Mass Conversion: Contour Graphs

Contour graphs depicting the time needed to convert 50% of cloud mass to rain mass are shown for simulations with an initial mean cloud radius ($\bar{x}_c$) of $10 \text{ }\mu\text{m}$ (polluted) and $16 \mu\text{m}$ (clean). The polluted simulation requires a cloud self-collection phase to grow the mean cloud droplet size large enough for auto conversion to have a significant effect. Therefore, the cloud self-collection parameter can be expected to be strong and the auto conversion parameter to be weaker as is shown by the location of the thin-lined black box in the upper corner of Figure 4.4a.

Both graphs in Figure 4.4 are blow-ups of the graphs in Figure 4.2. In

**Figure 4.4: Contour Plot on μ_{1n} - μ_{1m} plane:
50% Conversion time, Polluted Cloud**

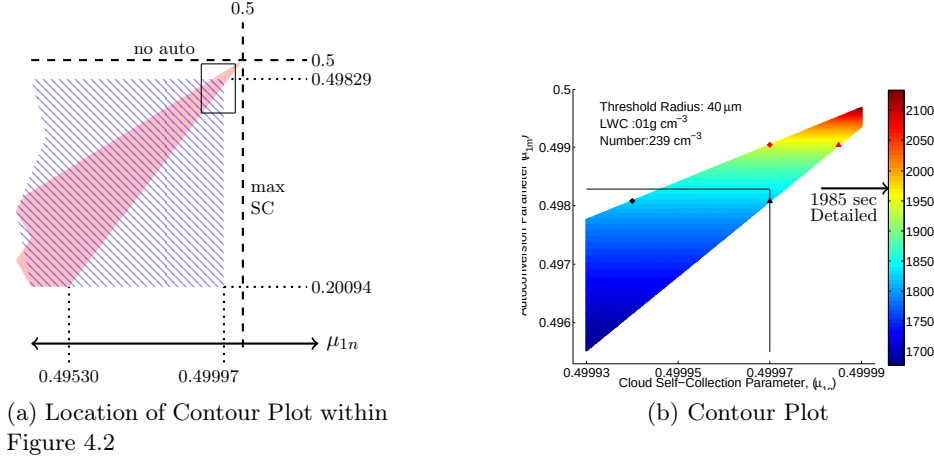
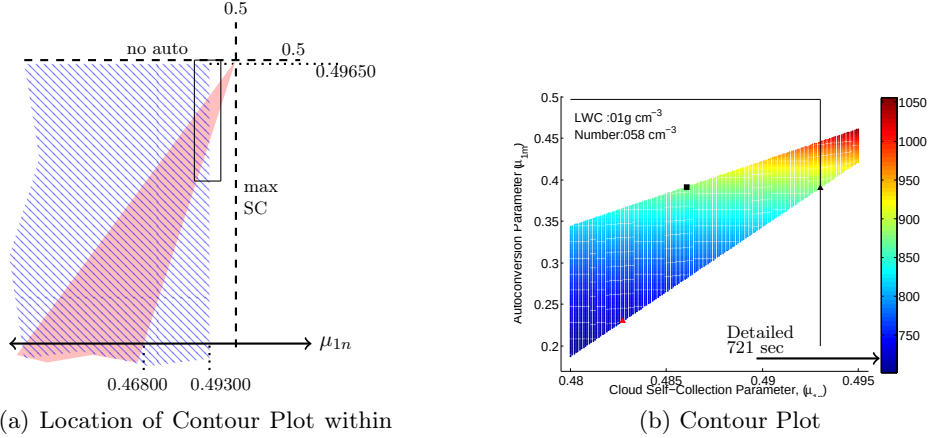


Figure 4.4: The graph (a) is a blow-up of the two graphs in Figure 4.2. The solid (red) region contains values of the cloud self-collection and auto conversion parameters that adhere to consistency of number and conservation of mass. The patterned (blue) region contains values which reproduce Siefert and Beheng's bulk parameterization. The thin box in the upper right is the region of the contour graph (b), which shows that instant at which rain mass exceeds cloud mass as a function of the two stochastic parameters. The solid lines in the contour plot correspond to the upper right edge of the patterned (blue) region in (a). These graphs are representative of the situation when the threshold radius is $40 \mu\text{m}$ and the mean cloud radius is $10 \mu\text{m}$. The orientation of the intersection is given by $x^*/\bar{x}_c = 64$. Neither graphs here are to the same scale as Figure 4.2.

the graph on the left, the solid (red) region contains values of the cloud self-collection and auto conversion parameters that adhere to consistency of number and conservation of mass. The patterned (blue) region contains values which reproduce Siefert and Beheng's bulk parameterization. This graph is not to scale, but identifies where the contour map is located relative to the graphs in Figure 4.2. The two black line segments in the contour map correspond to the edge of the patterned (blue) area within the black box in the graph on the left. Stochastic parameter values below and to the left of the black lines will reproduce Seifert and Beheng's bulk parameterization given the bounds on their parameters $0 \leq \nu \leq 3$ and $0 \leq \tau \leq 1$. Stochastic parameter values which are above and to the right of the black lines do not reproduce their bulk parameterization.

**Figure 4.5: Contour Plot on μ_{1n} - μ_{1m} plane:
50% Conversion time, Clean Cloud**



(a) Location of Contour Plot within
Figure 4.3

(b) Contour Plot

Figure 4.5: These are the same as in Figure 4.4 except the graph on the left is a blow-up of the two graphs in Figure 4.3 and the mean cloud radius is $16\mu\text{m}$. The details related to the diamonds and square are explained in the text. The orientation of the triangle is given by $\frac{x^*}{\bar{x}_c} = 15.625$. Neither graphs in Figure 4.5 are to scale.

The contour map in Figure 4.4b shows how strongly the evolution is affected by small changes in the auto conversion parameter in this small corner in the region of permissible values. The black triangle identifies the stochastic parameter values (0.49997, 0.49808) and the red diamond is (0.49997, 0.49904). These parameters convert 50% of cloud droplet mass to rain droplet mass in 1824 and 1944 seconds, respectively. The black diamond and the red triangle have the same 50% mass conversion time as the corresponding marks of the same colour. This is depicted in the contour graph by noting that contour lines are horizontal, i.e. mass conversion by the polynomial kernel is independent of the cloud self-collection parameter. The evolution curves corresponding to these parameters are in Figure 4.8.

The red marks are located outside of the region of stochastic parameter values that will reproduce Seifert and Beheng's bulk parameterization. The stochastic parameters are constant throughout the evolution. Seifert and Beheng's non-dimensional time parameter τ changes as the cloud matures. The auto conversion parameter μ_{1m} can be considered as an average of possi-

ble auto conversion parameter values during the maturation of a cloud. The τ function $\Phi_{\text{auto}}(\tau)$ is concave. Jensen's inequality shows that the convex function of a mean gives an underestimate (Larson et. al. 2001). We claim that the concave function gives an overestimate, and thus unduly increases the strength of the auto conversion parameter. When the incidental strengthening of the auto conversion parameter is mitigated by relaxing the restrictions imposed by the bounds of ν and τ on the values of the stochastic parameters, the stochastic model produces results consistent with detailed results using stochastic parameters identified by the (red) diamond.

**Figure 4.6: Contour Plot on μ_{1n} - μ_{1m} plane:
Mean Rain Radius, at the 50% conversion time**

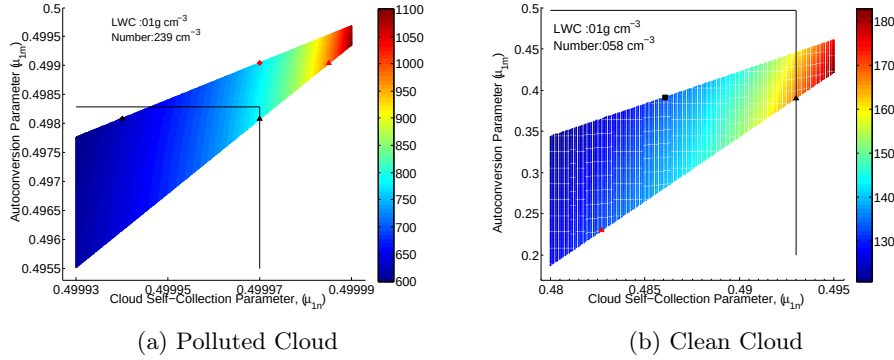


Figure 4.6: Mean rain radius at the time of 50% mass conversion to rain droplets. Initial conditions: 1 g m^{-3} for both, and 239 ($16 \mu\text{m}$) and 58.3 droplets cm^{-3} ($16 \mu\text{m}$) initial droplet concentration for (a) and (b), respectively.

The contour graph for simulations with an initial mean cloud radius of $16 \mu\text{m}$ is shown in Figure 4.5b with the thin black lines giving the boundary of the ‘SB-subset.’ The solid (red) and patterned (blue) regions serving the same purpose as in Figure 4.4. These graphs detail the regions presented in Figure 4.3. The black triangle, and square, identify the stochastic parameter pairs $(0.493013, 0.390833)$ and $(0.486060, 0.391093)$, respectively. These each convert 50% of mixing ratio to rain mixing ratio in 880 seconds. The evolution curves of both pairs are shown in Figure 4.9. The red diamond identifies the stochastic parameter values $(0.482682, 0.229417)$ and converts 50% of mixing ratio to rain mixing ratio in 726 seconds.. A cloud with a mean cloud radius of $16 \mu\text{m}$ has more liquid water content contained in larger cloud droplets. When the mean cloud radius is larger, say $16 \mu\text{m}$, stronger auto conversion process (shown by a smaller valued parameter) is expected.

This is seen when comparing the results from the simulations with an initial mean droplet radius of $10\mu\text{m}$.

Mean Rain Radius: Contour Graphs

The mean rain radius is an important complement to the 50% conversion time metric in a zero-degree parcel cloud model. The 50% conversion time quantifies how quickly cloud water is redistributed from being contained in cloud droplets to forming rain droplets. However, in a parcel model simulation the newly formed rain droplets are essentially “still in the cloud,” and have not started precipitating. This is a significant difference between a parcel model and a 1-D rain shaft model. By analyzing the conversion time in conjunction with mean rain droplet radius at 50% conversion time, we can get an idea of the onset of precipitation for particular stochastic parameter values relative to other stochastic parameter values and relative to other bulk and detailed models. A 1-D model would be appropriate when comparing to observational data. However, the inclusion of observational data is beyond the scope of this work.

**Figure 4.7: Contour Plot on μ_{1n} - μ_{1m} plane:
Mean Rain Radius, after the first time step**

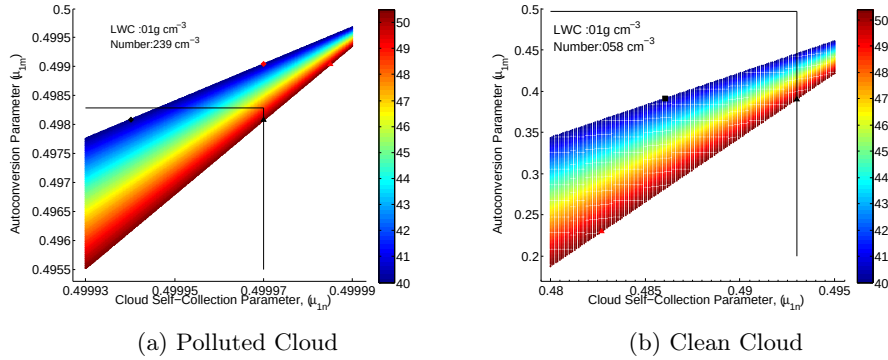


Figure 4.7: Mean rain radius after the first time step. Initial conditions: 1 g m^{-3} for both, and 239 ($16\mu\text{m}$) and 58.3 droplets cm^{-3} ($16\mu\text{m}$) initial droplet concentration for (a) and (b), respectively.

A larger mean rain droplet radius at the time of 50% conversion will lead to a quicker onset of precipitation, and a smaller mean rain droplet radius at that time indicates a slower onset of precipitation. The mean rain radius

at the 50% time is shown in Figure 4.6a and 4.6b for both the polluted ($10 \mu\text{m}$) case and the clean ($16 \mu\text{m}$) case, respectively. The boundary of the ‘SB subset region’ and the same triangular and square markers, identifying the location of particular parameter values, as shown in Figures 4.4 and 4.5 are replicated in the rain radius contour plot. These two contour plots also show that there is a greater dependence, of the mean rain radius at the 50% time of conversion, on the cloud self-collection parameter than on the auto conversion parameter.

Figure 4.8: Evolution Curves: Polluted Cloud

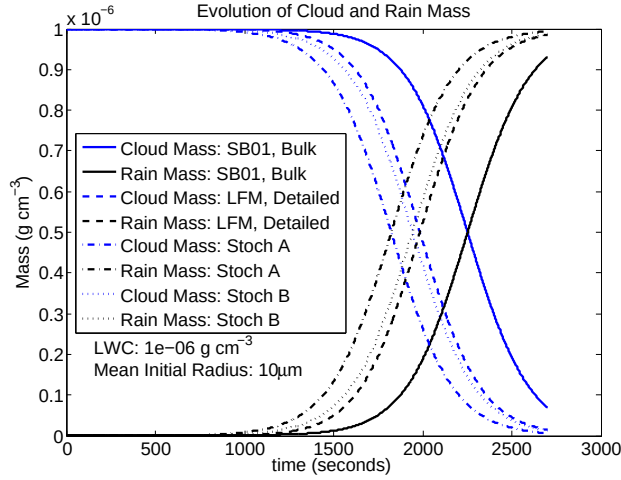


Figure 4.8: The stochastic bulk parameterization is evolved using the parameters listed in the graph. The parameter values are high lighted in Table 4.3. Initial conditions: 1 g m^{-3} , $238.7 \text{ droplets cm}^{-3}$, and $10 \mu\text{m}$ mean radius. The evolution is compared to a detailed evolution of the kinetic collection equation using Bott’s Linear Flux Method (1998) and Seifert and Beheng’s bulk parameterization. The stochastic parameterization evolves the aggregate mass faster than both Seifert and Beheng’s and the detailed results, and is closer to the detailed results than Seifert and Beheng’s parameterization. The parameters for Stoch A are $\mu_{1n} = 0.49997$, and $\mu_{1m} = 0.49808$, and for Stoch B: $\mu_{1n} = 0.49997$, and $\mu_{1m} = 0.49904$.

The mean rain radius at the first time step is shown in the two contour plots in Figure 4.7. In both cases, the upper boundary has an mean rain radius at the first time step of $40 \mu\text{m}$ which is equal to the threshold radius. This boundary separates the pre-collision droplets that form a rain droplet during the auto conversion process from ones that form a cloud droplet during this particular collision process. Since the latter contradicts the definition of auto conversion, this upper boundary separates physically allowable stochastic pa-

parameter values from ones that are not allowable. At the lower boundary, the mean rain radius at the first time step is $50.4 \mu\text{m}$. This boundary separates the pre-collision cloud droplets that form a rain droplet during the auto conversion process from ones that formed by pairs of rain droplets during this particular collision process. Since the latter contradicts the definition of auto conversion, this lower boundary also separates physically allowable stochastic parameter values from ones that are not allowable. The range of rain radii at the first time step is important when using particular turbulent kernels in the stochastic bulk model.

For instance, a tabulated turbulent kernel by Pinsky et. al. (2008) has the greatest enhancement when smaller cloud droplets are coalescing with larger cloud droplets as can be seen in a contour plot of their collision kernel that contains the strongest turbulence ($1000 \text{ TKE cm}^2\text{s}^{-3}$ and $20,000 \text{ R}$), the graph on the right in Figure 2.2. A turbulent kernel such as this would be best served by stochastic parameters along the upper boundary ($\overline{x_r}(t=1) = 40\mu\text{m}$) since the strongest enhancement of collisions occurs with the smallest of droplets, the predominate size of newly produced rain droplets be approximately equal to the size of the threshold droplet: $x^* = 40\mu\text{m}$.

In contrast, a parameterized kernel by (Franklin 2008) has stronger enhancement factors when droplets of nearly the same size coalesce and the greatest enhancements are when the largest of the cloud droplets coalesce with each other as can be seen in a contour plot of her collision kernel that contains the strongest turbulence ($50.5 \text{ TKE cm}^2\text{s}^{-3}$ and 1500 R), the graph on the left in Figure 2.2. This type of turbulent kernel would be best represented by the stochastic bulk model when using parameters along the lower boundary ($\overline{x_r}(t=1) = 50.4\mu\text{m}$). Two cloud droplets with radii slightly less than that of the threshold droplet would produce a rain droplet with a radii of $50.4 = (2 * 40^3)^{(1/3)}$.

Evolution of Mass Conversion

Cloud and rain mixing ratios are evolved using three methods: (i) the new ‘SDE-based’ stochastic bulk parameterization, (ii) the bulk parameterization by Seifert and Beheng (2001), and (iii) a detailed scheme using Bott’s Linear Flux Method (LFM) from 1998. The results from the detailed method are taken as the benchmark. Mixing ratios in polluted clouds are evolved for 45 minutes, and clean clouds (larger initial mean droplets) for 30 minutes. The stochastic parameter pairs chosen for these simulations are taken from the

marker locations on the contour plots in Figures 4.4 and 4.5.

Table 4.3: Evolution Results: Polluted Cloud

(μ_{1n}, μ_{1m})	$q_c = q_r$		$t = 1$ second
	seconds	$\bar{x}_r$ (μm)	$\bar{x}_r$ (μm)
(0.49994008, 0.49808267)	1824	628.2	40.0
(0.49997003, 0.49808213)	1824	791.5	50.4
(0.49997003, 0.49904107)	1944	773.9	40.0
(0.49998501, 0.49904053)	1944	975.2	50.4
Bulk: Seifert & Beheng (2001)	2253	151.3	39.6
Detailed: Linear Flux Method	1985		

Table 4.3: The first column gives the parameter values used to acquire the data in the remaining columns. The second and third columns give the time (seconds) and the mean rain radius (μm) at the first time step that rain mixing ratio exceeds cloud mixing ratio. The fourth column gives the mean rain radius at the first time step. The last column gives the time (seconds) at which the mean rain radius first equals the threshold radius. The four parameter pairs correspond to the four markers in Figure 4.4.

For the polluted cloud case, the stochastic parameter pairs used to generate the two stochastic curves in Figure 4.8 are in the second and third rows of Table 4.3. The first and fourth rows show simulations that generate the same 50% conversion time, but have different mean rain radii at the 50% time. The stochastic parameters that produce a 50% conversion time of 1944 seconds were specifically chosen to match the detailed results of 1985 seconds. Parameters that exactly reproduced the time of 1985 seconds were available; however, these evolution curves were indistinguishable. So, we chose values that highlighted how closely the curves matched. Seifert and Beheng’s conversion rate was considerably slower. However, the retarded growth by Seifert and Beheng’s parameterization is consistent with Franklin’s (2008) analysis that their model of auto conversion underestimates the auto conversion rate (Franklin 2008).

Referring to Figure 4.4, we see that the intersection of the ‘SB-subset’ region and the ‘region of validity’ is a truncation of the triangle due to the limits im-

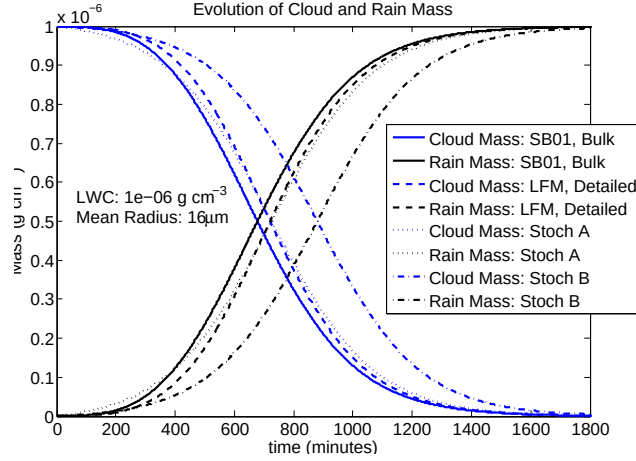
Figure 4.9: Evolution Curves: Clean Cloud

Figure 4.9: Same as Figure 4.8 except for the initial conditions: 1 g m^{-3} , $58.3 \text{ droplets cm}^{-3}$, $16 \text{ } \mu\text{m}$ mean radius. The cloud self-collection and auto conversion parameters for Stoch A are $\mu_{1n} = 0.482682$, and $\mu_{1m} = 0.229417$ and for Stoch B are $\mu_{1n} = 0.493013$, and $\mu_{1m} = 0.390833$. The latter pair generate an evolution curve that adhere to all physical constraints and lies within the region generated by the bound of Seifert and Beheng's parameters ν and τ .

posed by the values of ν and τ that Seifert and Beheng chose. When recreating their model with the stochastic parameterization, the space of stochastic parameter values was created using the maximums and minimums of ν and τ . However, τ changed during the evolution while the stochastic parameters remained constant. This arbitrary limitation calls for the relaxation of the truncation on the triangle due to the bounds on μ_{1n} and μ_{1m} imposed by ν and τ because the τ function is concave. By applying a principle similar to Jensen's inequality (Larson et. al. 2001), the overestimation of the auto conversation terms are mitigated. To compensate for this acceleration, values of μ_{1n} and μ_{1m} which inhibit the evolution are allowed. These values are stochastic parameter pairs of (μ_{1n}, μ_{1m}) , such that $\mu_{1n} > 0.49997$ and $\mu_{1m} > 0.49829$ are allowed.

Choosing stochastic parameters that adhere to the threshold droplet restriction, i.e. contained within the (solid, red) triangle in Figure 4.2, but not necessarily adhering to the derived restrictions on the auto conversion parameter, produces a bulk evolution curve more reflective of the detailed benchmark as shown in Figure 4.8. Figure 4.4 plots the locations of the

coordinates (μ_{1n}, μ_{1m}) used to generate the results of the mean stochastic parameterization in Figure 4.8 relative to the linear boundaries given by the relationship of auto conversion and the droplet threshold.

Table 4.4: Evolution Results: Clean Cloud

(μ_{1n}, μ_{1m})	$q_c = q_r$		$t = 1 \text{ second}$
	seconds	$\bar{x}_r \text{ } (\mu\text{m})$	$\bar{x}_r \text{ } (\mu\text{m})$
(0.482682, 0.229417)	726	132.4	50.4
(0.493013, 0.390833)	880	167.4	50.4
(0.486060, 0.391093)	880	132.9	40.0
Bulk: Seifert & Beheng (2001)	678	72.3	39.6
Detailed: Linear Flux Method	721		

Table 4.4: The first column gives the parameter values used to acquire the data in the remaining columns. The second and third columns give the time (seconds) and the mean rain radius (μm) at the first time step that rain mixing ratio exceeds cloud mixing ratio. The fourth column gives the mean rain radius at the first time step. The last column gives the time (seconds) at which the mean rain radius first equals the threshold radius. The three parameter pairs correspond to the three markers in Figure 4.5

For the clean cloud case, the stochastic parameter pairs used to generate the two stochastic curves in Figure 4.9 are in the first two rows of Table 4.4. The evolution curve, ‘Stoch A,’ is detailed in the first row of Table 4.4 and closely matches the 50% conversion time of the detailed simulation. For the initial 5-7 minutes the conversion of cloud water to rain water is excessive. At approximately 400 seconds the rate of conversion, as indicated by the slope of the respective graphs, is less than the rate for the detailed simulation. The slower conversion rate of the stochastic parameterization persists for the duration of the evolution.

When $\bar{x}_c = 16\mu\text{m}$ cloud mass is closer to the cloud-rain threshold. To accurately model this initial condition a more aggressive auto conversion parameter is needed. Consequently, values of the auto conversion parameter μ_{1m} need to be smaller and are situated more towards the centre of the quadrilateral region. The preferred stochastic value relative the region on which one can take values is shown in Figure 4.5. This is in contrast to a

more polluted initial condition such as when $\bar{x}_c = 10\mu\text{m}$. In this polluted situation the cloud mass is further from the cloud-rain threshold and is more accurately modelled by a weaker auto conversion parameter, and weaker to the extent that the concavity bias of Seifert and Beheng’s non-dimensional time parameter places an excessive restriction on the cloud self-collection and auto conversion stochastic parameters as is shown by the diamond (red) marker in Figure 4.4 being outside the patterned (blue) region.

4.3.3 Sensitivity Tests

Two types of sensitivity tests were performed. The effect that the time step had on the time required to convert 50% of cloud droplets to rain droplets and the rain droplet radii at that time. The third sensitivity test examines the effect that the rain number parameter has on these metrics. Both tests were performed using an initial mean cloud radius of $10\mu\text{m}$, and cloud self-collection and auto conversion parameters of $(\mu_{1n}, \mu_{1m}) = (0.49997, 0.49808)$.

Table 4.5: Sensitivity: Rain Self-Collection Parameter

μ_{4np}	$q_c = q_r$	
	seconds	$\bar{x}_r$ (μm)
0.40	1824	742.7
0.30	1824	775.0
0.25	1824	791.6
0.20	1824	808.5
0.10	1824	843.3

Table 4.5: Varying the rain self-collection parameter show little effect on rain droplet size. When μ_{4np} varies from 0.10 to 0.40, the mean rain radius at the time that half of the liquid water content is converted to rain varies by $100.6\mu\text{m}$ (12.7%).

As expected, the rain number stochastic parameter μ_{4np} only effects the evolution of rain number. When μ_{4np} is varied from 0.10 to 0.40, the mean rain radius varies by 12.7% according to Table 4.5.

Varying the time step from 0.1 seconds to 30 seconds has a negligible effect

on the time required to convert half of the initial cloud mass to rain droplets (column two in Table 4.6). The mean rain radius at the time that half of the liquid water content is converted to rain varies by $4.9\mu\text{m}$ (0.62%) between time steps of 0.1 and 30 seconds. No advantage is gained by reducing the time step smaller than the 1 second used in the simulations in this paper. It may be possible to relax the time step to 30 seconds without significantly compromising accuracy. It should be noted that at 50 minutes of evolution the mean rain radius was greater than half a centimetre indicating that the omission of rain removal and droplet breakup were effecting the results.

Table 4.6: Sensitivity: Time Step

time step		$q_c = q_r$
seconds	seconds	$\bar{x}_r$ (μm)
120	1920	797.6
60	1860	818.0
30	1830	795.9
10	1830	800.1
1	1824	791.6
0.1	1823.6	791.0

Table 4.6: Varying the time step has a negligible effect on the time when rain mass exceeds cloud or the mean rain mass at that instant. The mean rain radius at the 50% time varies by only $4.9\mu\text{m}$ (0.62%) between 0.1 and 30 seconds.

4.4 Summary

We used the relationship of auto conversion to threshold droplet mass to define ‘regions of validity’ for the auto conversion (μ_{1m}) and the cloud self-selection (μ_{1n}) parameters such that the selection of values for these parameters would cause the auto conversion terms in the stochastic bulk model to produce a rain droplet when two cloud droplets coalesced. This restriction of the auto conversion parameter was only possible because each of the four occurrences of an auto conversion term in the stochastic bulk model was

derived independently of every other auto conversion term. To the best of the author’s knowledge, no other bulk cloud microphysics model of collision and coalescence was able to accomplish this physics-based restriction on the auto conversion term because every other bulk model only derived one auto conversion term and then made the other three auto conversion terms to be functions of the derived term.

We then applied an analytic collision kernel to the stochastic bulk model. This same collision kernel was used by Seifert and Beheng (2001) to derive a bulk model of collision and coalescence. Their bulk model is restricted to this kernel, while ours retains the flexibility to use a variety of collision kernels. Both models had precisely the same dependence on cloud and rain mixing ratio and number concentration. We matched the models term for term and solved for the stochastic parameters μ_{1m} and μ_{1n} as functions of their ad-hoc parameter. We defined an ‘SB subset region’ in the 2-D (μ_{1m}, μ_{1n}) space by applying the bounds that they gave for their parameters.

The intersection of the ‘region of validity’ and the ‘SB subset region’ defines a set of values for the stochastic parameters of cloud self-collection and auto conversion that both adheres to physical constraints and reproduces the bulk model by Seifert and Beheng (2001). We validated the stochastic bulk model by finding stochastic parameter values that very closely reproduced the 50% mass conversion time and the evolution curves. This was done for the cases of a polluted cloud (239 cm^{-3}) and a clean cloud (58.3 cm^{-3}). The model was further validated for both cloud types by noting that the mean rain radius at the first time step had physically meaningful bounds related to the process of auto conversion, i.e. the bounds on the rain radius at the first time step was dependent on the fact that process of auto conversion produces one rain droplet from two cloud droplets.

We collected data on the mean rain radius at the time of 50% mass conversion to rain, and related the upper ($50.4 \text{ }\mu\text{m}$) and lower bounds ($x^*=40 \text{ }\mu\text{m}$) of this radius to macroscopic cloud properties i.e. larger scale air movements that effect droplet sizes differently. Each bound was associated with a different type of turbulent collision kernel which will be developed further in Chapter 5.

Lastly, sensitivity tests showed that for this kernel, the rain self-collection parameter has no effect on the evolution time and a small effect on the rain droplet radius. The time step can be extended to 30 seconds with no noticeable loss of accuracy, and a time step as short as 0.1 seconds did not improve performance.

5 | Extension

to Hydrodynamic and Turbulent Kernels

The Stochastic Bulk Model of collision and coalescence is extended to include hydrodynamic and turbulent kernels. Simulations for each of these three realistic kernels are performed for a polluted cloud and a clean cloud. Polluted, and clean, clouds have number concentrations of 239 cm^{-3} and 58.3 cm^{-3} , respectively, yielding a total of six simulations. All six simulations use a liquid water content of 1 g cm^{-3} . Therefore the initial mean cloud radius in polluted, and clean, clouds are $10 \mu\text{m}$, and $16 \mu\text{m}$, respectively. All simulations use a rain number parameter (μ_{4np}) of 0.25. Unless otherwise noted, all simulations ran for one hour and if 50% did not occur by that time then the value of 3600 seconds was assigned for the purposes of the contour plots.

For each of the six simulations, results are presented in five forms: (i) a contour plot showing the time required to convert 50% of cloud mass to rain mass as a function of the cloud self-collection and auto conversion parameters with chosen parameter pairs highlighted, (ii) a table giving statistics associated with the highlighted parameter pairs, (iii) evolution graphs produced by these parameter pairs compared to a detailed evolution, (iv) contour maps showing the mean rain radius at the first time step and at the time of 50% conversion, and (v) four plots showing time and mean rain radius at the time of 50% conversion as a function stochastic parameters (a slice of the respective contour plots).

5.1 Hydrodynamic Kernel

The hydrodynamic kernel used in these simulation is detailed in Section 2.2.2 and presented in the tables in Section A.1. Collision rates from this kernel are the base rates that the turbulent kernels in Sections 5.2 and 5.3 enhance.

5.1.1 Polluted Cloud: $239 \text{ cm}^3\text{s}^{-1}$

In Figure 5.1 , we show the contour plot of the 50% conversion time as a function of the parameters μ_m and μ_n within the “region of validity” as discussed in Chapter 4. The 50% conversion time for the detailed method (2359sec) and Khairoutdinov and Kogan’s (1966sec) deterministic bulk model on the colour bar. Although the minimum value for μ_{1n} is 0.3, the range of allowable values extends to -0.5. The region not shown has small variation and is omitted to detail the region of interest. The red marks locate stochastic parameter pairs that approximate the 50% conversion time for the detailed method. The black marks do the same for approximating the deterministic model of Khairoutdinov and Kogan (2000).

The triangular marks give stochastic parameters that produce a mean rain radius of $50.4 \mu\text{m}$ at the first time step. This would be the result of the largest cloud droplets, nearest in size but just smaller than the threshold radius, colliding to produce a rain droplet. The square marks give stochastic parameters that produce a mean rain radius of $40.0 \mu\text{m}$ at the first time step. This would be the result of the smallest cloud droplets colliding to produce a rain droplet that is the same size as the threshold droplet.

The 50% conversion time using the hydrodynamic kernel has very little dependence on the cloud self-collection (μ_{1n}) parameter. There is a slight dependence when the mean rain radius at the first time step is $40.0 \mu\text{m}$. Along the upper edge of the triangle we see the 50% conversion time being slightly delayed when the cloud self-collection parameter is weaker.

The four pairs of stochastic parameters marked on Figure 5.1 are detailed in Table 5.1. As expected, a larger mean rain radius at the first time step produces a larger mean rain radius at the time of 50% conversion, which is seen when comparing the first two rows and when comparing the last two rows. Also, the parameter pairs in the first two rows produce a nearly

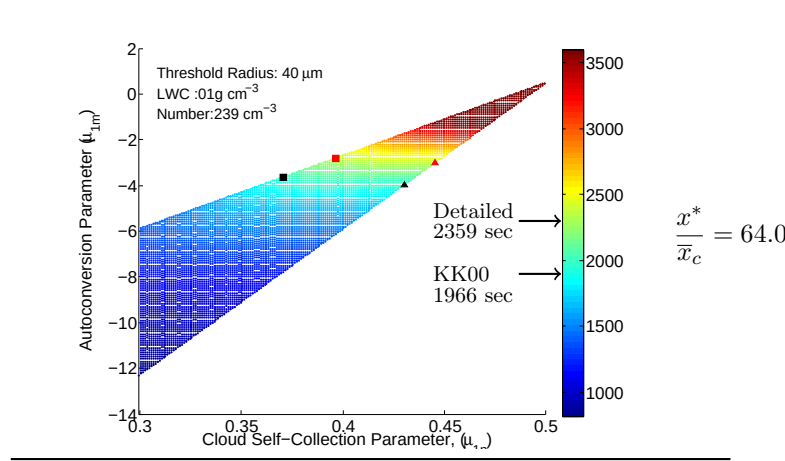
Figure 5.1: Hydrodynamic, Polluted: Contour, 50% conversion

Figure 5.1: Contours of 50% conversion time as a function of the stochastic parameters μ_{1m} and μ_{1n} . The red, and black, triangles locate the stochastic parameters: (0.44527, -3.000) and (0.43035, -3.957), respectively. The red, and black, marks indicate the location of stochastic parameters that converts 50% of the cloud mixing ratio to rain mixing ratio in 2366 seconds and 1975 seconds, respectively.

equal 50% conversion time despite the auto conversion parameter varying by $\approx 8.4\%$.

Table 5.1: Hydrodynamic, Polluted: Stochastic Parameters

(μ_{1n}, μ_{1m})	$q_c = q_r$		$t = 1$ second
	seconds	$\bar{x}_r$ (μm)	$\bar{x}_r$ (μm)
(0.37065, -3.6392)	1976	55.1	40.0
(0.43035, -3.9576)	1975	205.1	50.4
(0.39652, -2.8114)	2354	176.3	40.0
(0.44527, -3.0000)	2366	226.1	50.4

Table 5.1: The first column gives the parameter values used to acquire the data in the remaining columns. The second and third columns give the time (seconds) and the mean rain radius (μm) at the first time step that rain mixing ratio exceeds cloud mixing ratio. The fourth column gives the mean rain radius at the first time step.

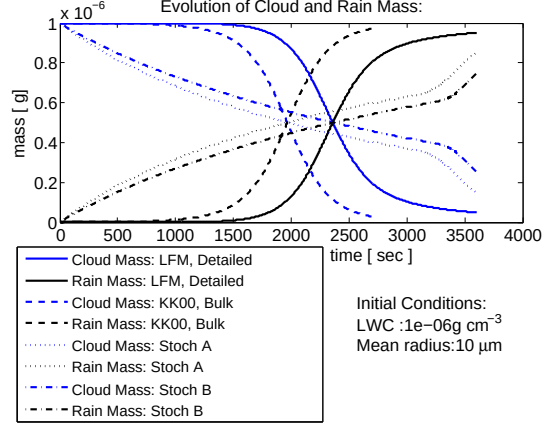
Figure 5.2: Hydrodynamic, Polluted: Evolution Curves

Figure 5.2: Bulk evolution of mass conversion comparing the stochastic model with the Khairoutdinov and Kogan’s deterministic model benchmarked against a detailed method for a polluted cloud with an initial mean droplet radius of $10\mu\text{m}$.

The two stochastic curves (Stoch A and Stoch B) in Figure 5.2 correspond to rows 2 and four in Table 5.1, respectively. They also correspond to the black and red triangles in Figure 5.1, respectively. These two stochastic evolution curves are compared to curves produced by Bott’s LFM detailed method and by Khairoutdinov and Kogan’s bulk method. We see a stark difference in the evolution curves between the deterministic ones and the stochastic ones. The stochastic ones have a near linear conversion rate until the conversion accelerates at approximately 3200 seconds and 3400 seconds for Stoch A and Stoch B, respectively. A more complex model that includes nucleation and evaporation, and that is compared with observational data, in a 1-D rainshaft may be useful to determine if this loss of cloud mass corresponds with cloud termination.

Parameters of the Stochastic curves in Figure 5.2:

Stoch A : $\mu_{1n} = 0.43035$, and $\mu_{1m} = -3.957$

Stoch B : $\mu_{1n} = 0.44527$, and $\mu_{1m} = -3.000$.

The stochastic evolution curves are strikingly different than the deterministic curves. Except for the last few minutes of the simulation, the conversion rate of cloud mass to rain mass is steady. This is in contrast to a slower early conversion and a faster conversion for the deterministic bulk model and the detailed simulation near 2000 and 2500 seconds, respectively. This

Figure 5.3: Hydrodynamic, Polluted: Mean Rain Radius

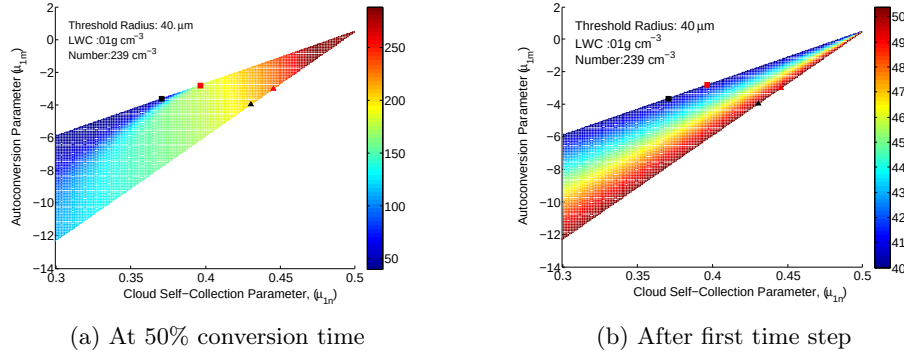


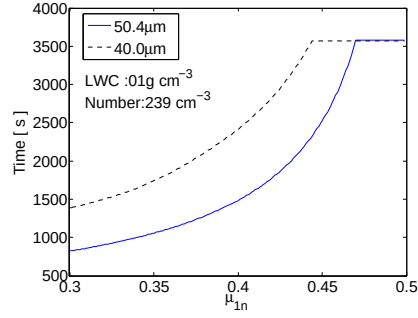
Figure 5.3: The mean rain droplet radius at the time of 50% mass conversion is graphed in (a). The mean rain droplet radius at the first time step as a function of the cloud-self-collection and auto conversion parameters for a polluted cloud with an initial mean droplet radius of $10\mu\text{m}$ is graphed in (b).

could be due to the mean of the stochastic processes being taken over the entire simulation rather than at times when physically meaningful phenomena occur, such as the collision and coalescence being dominated by cloud self collection, or by accretion, or by rain self collection. This needs further research.

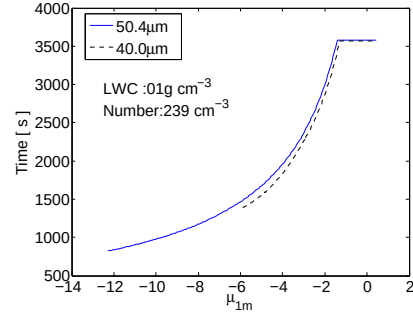
The mean rain droplet radius, both at the 50% conversion time and at the time of the first time step, are plotted as functions of the cloud self-collection and auto conversion parameters in Figure 5.3. The contour of the mean rain droplet radius at the 50% conversion time is plotted in Figure 5.3a. We see a strong dependence on the cloud self-collection parameter and a weaker dependence on the auto conversion parameter. This pattern is disrupted when the conversion time is delayed and then the mean rain radius severely restricted for simulations that begin by colliding the smallest cloud droplets in an auto conversion process.

The mean rain radius at the first time step, plotted in Figure 5.3b, increases in a straight-forward manner from $40\mu\text{m}$ to $50.4\mu\text{m}$. The maximum radius of a newly formed rain drop due to auto conversion is $50.4\mu\text{m}$ because the largest cloud droplets have radii of $40\mu\text{m}$ ($50.4 = (2 \cdot 40^3)^{1/3}$).

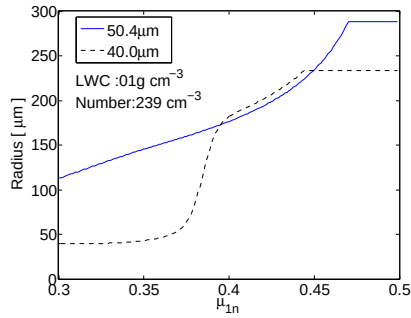
The two of the edges of the triangle in Figures 5.1 and 5.3 are detailed in Figure 5.4. The graphs (a) and (b) show the 50% conversion time as a function

Figure 5.4: Hydrodynamic, Polluted: Edge Plots

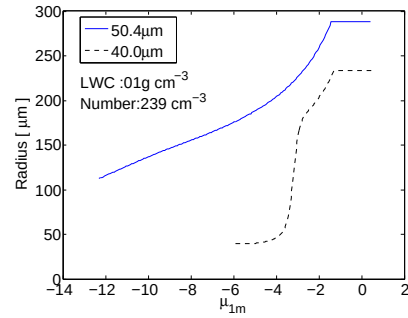
(a) 50% conversion time over cloud self-collection parameter



(b) 50% conversion time over auto conversion parameter



(c) Mean Rain Radius at 50% conversion time over cloud self-collection parameter



(d) Mean Rain Radius at 50% conversion time over auto conversion parameter

Figure 5.4: The graphs (a) and (b) show the time to convert 50% of mass to rain droplets as a function of the cloud self-collection, and auto conversion, parameter, respectively. The graphs (c) and (d) show the mean rain radius at the time that 50% of mass has been converted to rain droplets as a function of the cloud self-collection, and auto conversion, parameter, respectively. Each graph contains curves corresponding to the mean rain droplet radius at the first time step of $50.4\text{ }\mu\text{m}$ (blue) and $40\text{ }\mu\text{m}$ (black), i.e. along the upper edge and lower edge of the triangle in the respective contour maps.

of each parameter. The graphs (c) and (d) show the mean rain radius as a function of each parameter. Note that the upper edge of the triangle has a higher lower bound of values of μ_{1m} than the lower edge. Therefore, the black curves on the right two plots do not have the same domain as the blue curves.

Figure 5.5: Hydrodynamic, Clean: Contour, 50% conversion

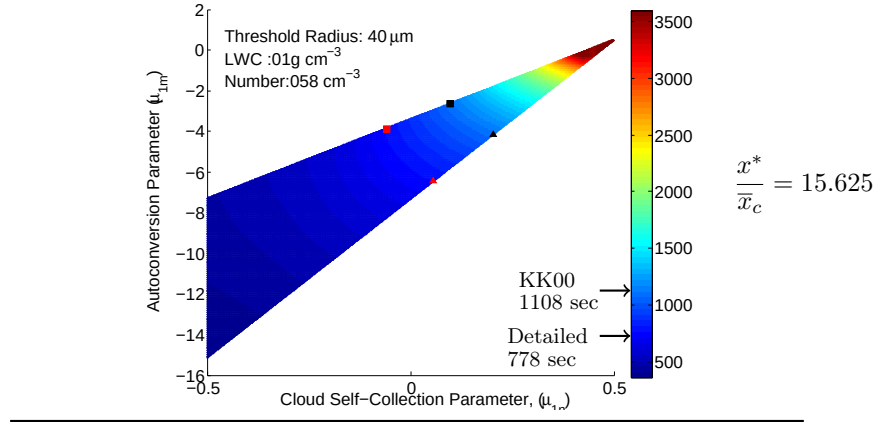


Figure 5.5: The red, and black, triangles locate the stochastic parameters: (0.05644, -6.430) and (0.21428, -3.964), respectively. The red, and black, marks indicate the location of stochastic parameters that converts 50% of the cloud mixing ratio to rain mixing ratio in 775 seconds and 1144 seconds, respectively.

All of the plots are monotonically increasing, and plateau at the simulation end time due to the construction of the contour plots. The 50% conversion time plots show a smooth curve until the 3600 second threshold is reached for each parameter plot. The rain radius plots show a smooth curve when the largest cloud droplets are consumed by auto conversion ($\bar{x}_r(t=1)=50.4$), but a delay in the growth of the mean rain radius for a weaker cloud self-collection process. A stronger auto conversion parameter increases the mean rain radius before a weaker cloud self-collection parameter will have the same effect.

5.1.2 Clean Cloud: $58.3 \text{ cm}^3\text{s}^{-1}$

In Figure 5.5 we plot the contours of the 50% conversion time obtained by the stochastic bulk model as the parameters μ_n and μ_m are varied. The 50% conversion time for the detailed method (778 sec) and Khairoutdinov and Kogan's (1108sec) deterministic bulk model are shown on the colour bar. Unlike the simulations for the polluted cloud, the clean cloud has an initial mean cloud radius that is closer to the threshold radius ($40\mu\text{m}$), and

the minimum value for μ_{1n} extends to -0.5. Again, the red marks locate stochastic parameter pairs that approximate the 50% conversion time for the detailed method. The black marks do the same for approximating the deterministic model.

Figure 5.2: Hydrodynamic, Clean: Stochastic Parameters

(μ_{1n}, μ_{1m})	$q_c = q_r$		$t = 1 \text{ second}$
	seconds	$\bar{x}_r \text{ } (\mu\text{m})$	$\bar{x}_r \text{ } (\mu\text{m})$
(-0.05845, -3.86283)	778	108.6	40.0
(0.05644, -6.43056)	775	138.8	50.4
(0.09641, -2.65310)	1107	159.9	40.0
(0.20229, -4.15159)	1105	171.2	50.4

Table 5.2: This table is the same as Table 5.1 except the data is for clean cloud.

As with the polluted cloud simulations, the triangular marks give stochastic parameters that produce a mean rain radius of $50.4 \mu\text{m}$ at the first time step. This would be the result of the largest cloud droplets, nearest in size but just smaller than the threshold radius, colliding to produce a rain droplet. The square marks give stochastic parameters that produce a mean rain radius of $40.0 \mu\text{m}$ at the first time step. This would be the result of the smallest cloud droplets colliding to produce a rain droplet that is the same size as the threshold droplet.

The coloured contours in Figure 5.5 show that both the cloud self-collection and auto conversion parameters have a qualitatively equivalent effect on the 50% evolution time.

The four pairs of stochastic parameters marked on Figure 5.5 are detailed in Table 5.2. As expected, a larger mean rain radius at the first time step produces a larger mean rain radius at the time of 50% conversion, which is seen when comparing the first two rows of Table 5.2 and when comparing the last two rows of the same table. The difference between the mean rain radius at the time of 50% conversion is less pronounced then in the polluted case, but the conversion time is also short, by over a factor of two.

The two stochastic curves (Stoch A and Stoch B) in Figure 5.6 correspond to rows 2 and four in Table 5.2, respectively. They also correspond to the black

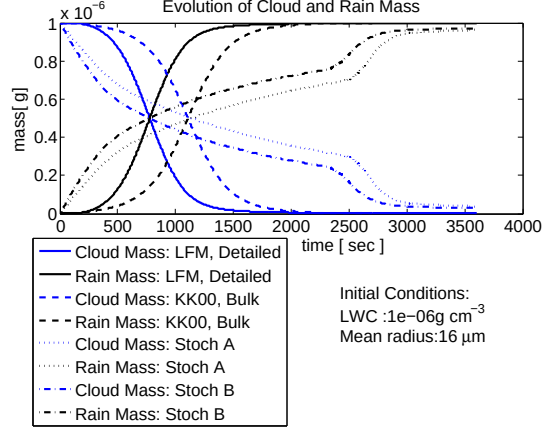
Figure 5.6: Hydrodynamic, Clean: Evolution Curves

Figure 5.6: Bulk evolution of mass conversion comparing the stochastic model with the Khairoutdinov and Kogan’s deterministic model benchmarked against a detailed method for a clean cloud with an initial mean droplet radius of $16 \mu\text{m}$.

and red triangles in Figure 5.5, respectively. These two stochastic evolution curves are compared to curves produced by Bott’s LFM detailed method and by Khairoutdinov and Kogan’s bulk method. We see a stark difference in the evolution curves between the deterministic ones and the stochastic ones. The stochastic ones begin with a strong conversion rate followed by a weaker rate until the conversion accelerates rapidly at approximately 2400 seconds and 2500 seconds for Stoch A and Stoch B, respectively. With respect to the sharp decline near 2400 and 2500 seconds, a more complex stochastic model that includes nucleation and evaporation, and that is compared with observational data, in a 1-D rainshaft may be useful to determine if this loss of cloud mass corresponds with cloud termination, thereby potentially avoiding errors in the detailed method that may (at least partially) cancel each other out, such as numerical diffusion and a midpoint approximation.

Parameters of the Stochastic curves in Figure 5.6:

Stoch A : $\mu_{1n} = 0.21428$, and $\mu_{1m} = -3.964$

Stoch B : $\mu_{1n} = 0.05644$, and $\mu_{1m} = -6.430$

As in the polluted case, the mean rain droplet radius, both at the 50% conversion time and at the first time step, are plotted as functions of the cloud self-collection and auto conversion parameters in Figures 5.7a and 5.7b, re-

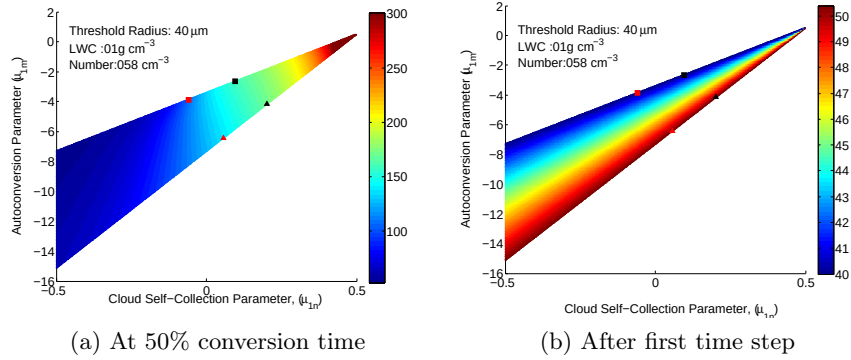
Figure 5.7: Hydrodynamic, Clean: Mean Rain Radius

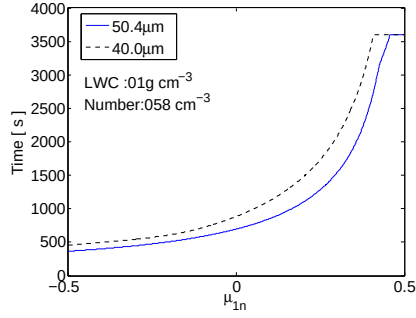
Figure 5.7: The mean rain droplet radius at the time of 50% mass conversion is graphed in (a). The mean rain radius at the first time step as a function of the cloud-self-collection and auto conversion parameters for a clean cloud with an initial mean droplet radius of $16\mu\text{m}$ is graphed in (b).

spectively. The contour of the mean rain droplet radius at the 50% conversion time is plotted in the graph on the left. We again see a strong dependence on the cloud self-collection parameter and a weaker dependence on the auto conversion parameter. This pattern is disrupted when the conversion time is delayed. However, the disruption is less pronounced than when the initial cloud radius is a smaller fraction of the threshold radius.

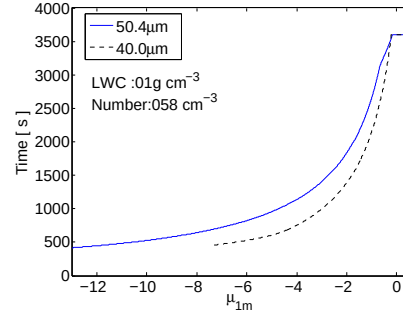
The graph in Figure 5.7b shows a smooth transition from the rain radius of 40 to $50\mu\text{m}$ at the first time step as was present in the polluted case.

The two of the edges of the triangle in Figures 5.5 and 5.7 are detailed in Figure 5.8. The graphs, Figure 5.8a and 5.8b, show the 50% conversion time as a function of each parameter. The graphs, Figure 5.8c and 5.8d, show the mean rain radius as a function of each parameter. Note that the upper edge of the triangle has a higher lower bound of values of μ_{1m} than the lower edge. Therefore, the black (dotted) curves on the right two plots do not have the same domain as the blue (solid) curves.

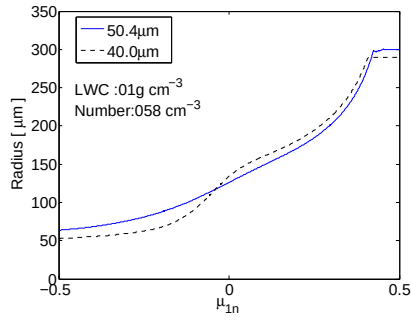
All of the plots are monotonically increasing, and plateau at the simulation end time due to the construction of the contour plots. The 50% conversion time plots show a smooth curve until the 3600 second threshold is reached for each parameter plot. The rain radius plots show a smooth curve when the largest cloud droplets are consumed by auto conversion ($\bar{x}_r(t=1)=50.4$).

Figure 5.8: Hydrodynamic, Clean: Edge Plots

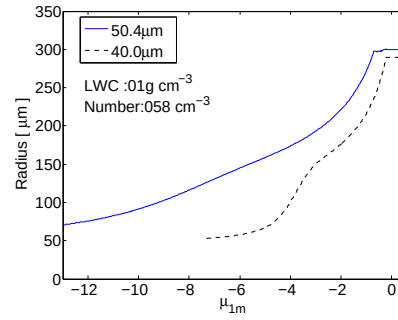
(a) 50% conversion time over cloud self-collection parameter



(b) 50% conversion time over auto conversion parameter



(c) Mean Rain Radius at 50% conversion time over cloud self-collection parameter



(d) Mean Rain Radius at 50% conversion time over auto conversion parameter

Figure 5.8: The same as in Figure 5.4 except that this is the hydrodynamic clean cloud case.

The dependence of rain radius at the time of 50% conversion is less smooth as a function of each parameter independently. The initial mean cloud radius is larger, and thus there is a smaller separation between this initial radius and the threshold radius. Possibly the cloud self-collection parameter and auto conversion parameters are ‘competing’ and producing an undulating (black) curve when the smaller cloud droplets dominate the auto conversion process w.r.t. the smoother (blue) curve, which is when the larger cloud droplets dominate the auto conversion process. The black curves in the rain radius (lower) plots of Figure 5.8 could be seen as a softened undulation compared to the same curves in Figure 5.4.

5.2 Parameterized Turbulent Kernel

The parameterized turbulent kernel used in this subsection is by Franklin et. al. (2007) and is further described in Section 2.2.3. The simulations performed in this section used turbulent values that are at the upper bounds of the valid range for their parameterization ($1500 \text{ cm}^2\text{s}^{-3}$ and 50.5 Reynolds number). The turbulent enhancement factor for this collision rate is shown in the left contour plot of Figure 2.2, and can be characterized as having a weaker typical large scale air flow in the cloud, but a stronger viscous dissipation rate. The contour graph indicates that the strongest enhancement occurs for droplets near the same size and when the droplets size is large enough such that deviations from Stokes drag is significant ($25\text{-}30 \mu\text{m}$). Coupling is the predominant turbulent phenomena for droplets with these characteristics (Wang et al. 1998, Pruppacher & Klett 1997).

Figure 5.9: Parameterized Polluted: Contour, 50% conversion

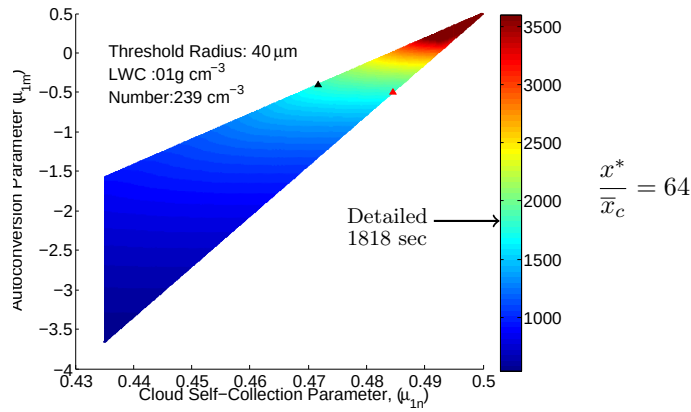


Figure 5.9: Contour plot of 50% conversion time for the stochastic bulk model applied to a parameterized turbulent kernel. The black, and red, triangles mark stochastic parameters: $(0.47175, -0.40421)$ and $(0.48452, -0.49000)$, respectively. When these parameters drive the bulk model, 50% of the cloud mixing ratio converts to rain mixing ratio in 1817, and 1806, seconds, respectively. The arrow points to the 50% conversion time for the detailed method.

5.2.1 Polluted Cloud: $239 \text{ cm}^3\text{s}^{-1}$

Figure 5.9 show the contours of the 50% conversion time for the stochastic bulk model as a function of the parameters μ_{1n} and μ_{1m} . The 50% conversion time for the detailed method (1818 sec) is shown on the colour bar. Unlike the simulations for the hydrodynamic kernel, the value for the cloud self-collection parameter is limited to those that are close to the maximum 0.5 which reduces the strength of the auto conversion process in the stochastic bulk model. Here, the red mark locates stochastic parameter pairs that approximate the 50% conversion time when the mean rain radius is $50.4 \mu\text{m}$ after the first time step. The black mark does the same for when mean rain radius is $40.0 \mu\text{m}$ after the first time step.

The coloured contours on Figure 5.9 show that the 50% conversion time has a stronger dependence on the auto conversion parameter and a weaker dependence on the cloud self-collection parameter except near the edge of the triangle where the mean rain radius is $40.0 \mu\text{m}$ at the first time step.

Table 5.3: Parameterized Polluted: Stochastic Parameters

(μ_{1n}, μ_{1m})	$q_c = q_r$		$t = 1 \text{ second}$
	seconds	$\bar{x}_r (\mu\text{m})$	$\bar{x}_r (\mu\text{m})$
(0.47175, -0.40421)	1817	124.3	40.0
(0.48452, -0.49000)	1806	210.5	50.4

Table 5.3: The same as Table 5.1 except the stochastic parameters are found in the contour map in Figure 5.9.

The two pairs of stochastic parameters marked by triangles (one red and one black) on Figure 5.9 are detailed in Table 5.3. As with the hydrodynamic case, a larger mean rain radius at the first time step produces a larger mean rain radius at the time of 50% conversion, which is seen when comparing the two rows. The difference between the mean rain radius at the time of 50% conversion is less pronounced then in the hydrodynamic case, but it should be noted that the values for both stochastic parameters lead to stronger cloud self-collection and auto conversion processes in the bulk model.

Parameters of the stochastic curves in Figure 5.10.

Stoch A : $\mu_{1n} = 0.48452$, and $\mu_{1m} = -0.49000$

Stoch B : $\mu_{1n} = 0.47175$, and $\mu_{1m} = -0.40421$

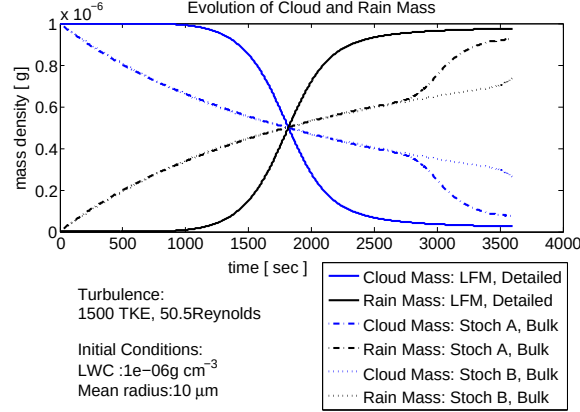
Figure 5.10: Parameterized, Polluted: Evolution Curves

Figure 5.10: Bulk evolution of mass conversion comparing the stochastic model benchmarked against a detailed method for a polluted cloud with an initial mean droplet radius of $10 \mu\text{m}$. The Stoch A curve is generated using $(\mu_{1n}, \mu_{1m}) = (0.48452, -0.49000)$. The Stoch B curve is generated using $(\mu_{1n}, \mu_{1m}) = (0.47175, -0.40421)$.

The two stochastic curves (Stoch A and Stoch B) in Figure 5.10 correspond to the two rows in Table 5.3, respectively. They also correspond to the black and red triangles in Figure 5.9, with a mean rain radius at the first time step of 40 and 50.4, respectively. Again, we see a stark difference in the evolution curves between the deterministic ones and the stochastic ones. As with the polluted hydrodynamic case, the stochastic ones have a near linear conversion rate until the conversion accelerates at approximately 2800 seconds and 3500 seconds for Stoch A and Stoch B, respectively.

The mean rain droplet radius, both at the 50% conversion time and at the first time step, are plotted as functions of the cloud self-collection and auto conversion parameters in Figures 5.11a and 5.11b. We see a strong dependence on the cloud self-collection parameter and a weaker dependence on the auto conversion parameter. This pattern is disrupted when the conversion time is delayed and then the mean rain radius severely restricted for simulations that begin by colliding the smallest cloud droplets in an auto conversion process. This is very similar to the polluted hydrodynamic case exempt the pattern occurs over a different range of stochastic parameter values.

The mean rain radius at the first time step (Figure 5.11b) shows the same

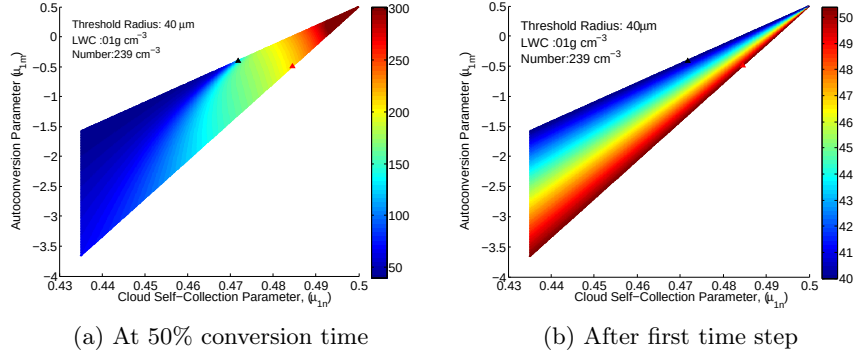
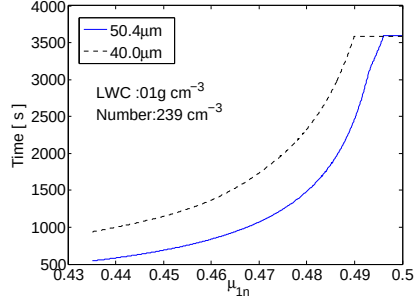
Figure 5.11: Parameterized, Polluted: Mean Rain Radius

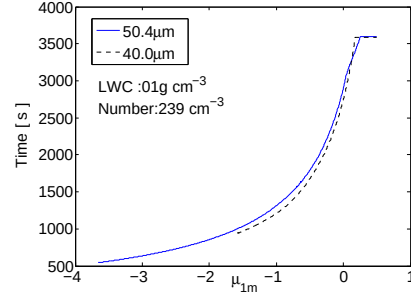
Figure 5.11: The mean rain droplet radius at the time of 50% mass conversion is graphed in (a). The mean rain droplet radius at the first time step as a function of the cloud-self-collection and auto conversion parameters for a polluted cloud with an initial mean droplet radius of $10 \mu\text{m}$ is graphed in (b).

pattern as both the clean and polluted hydrodynamic cases.

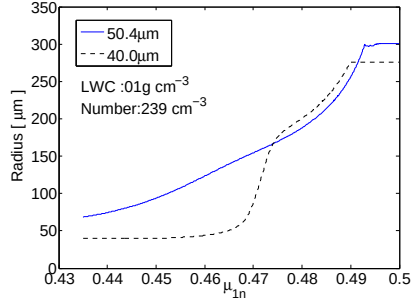
The two of the edges of the triangle in Figures 5.9 and 5.11 are detailed in Figure 5.12. These four graphs plot the same information as what were shown in each hydrodynamic case. They give very similar results to what were shown for the polluted hydrodynamic case in all four plots except that they range over different parameter values.

Figure 5.12: Parameterized, Polluted: Edge Plots

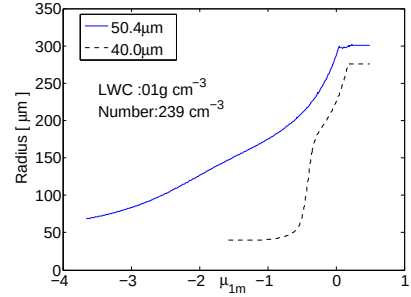
(a) 50% conversion time over cloud self-collection parameter



(b) 50% conversion time over auto conversion parameter



(c) Mean Rain Radius at 50% conversion time over cloud self-collection parameter



(d) Mean Rain Radius at 50% conversion time over auto conversion parameter

Figure 5.12: Same as Figure 5.4 except the bulk stochastic model uses Franklin's (2007) parameterized turbulent kernel.

5.2.2 Clean Cloud: $58.3 \text{ cm}^3 \text{ s}^{-1}$

Figure 5.13 shows the contours of the 50% conversion time for the stochastic bulk method as a function of the parameters μ_{1n} and μ_{1m} . The 50% conversion time for the detailed method (669 sec) is shown on the colour bar. Unlike the simulations for the polluted cloud, the clean cloud has an initial mean cloud radius that is closer to the threshold radius ($40 \mu\text{m}$). Also in contrast to the clean, hydrodynamic case the minimum value for μ_{1n} extends to 0.3, 40% of the range in the clean hydrodynamic case. Again, the red and black triangles locate stochastic parameter pairs that approximate the 50% conversion time for the detailed method given a mean rain radius at the first

Figure 5.13: Parameterized Clean: Contour, 50% conversion

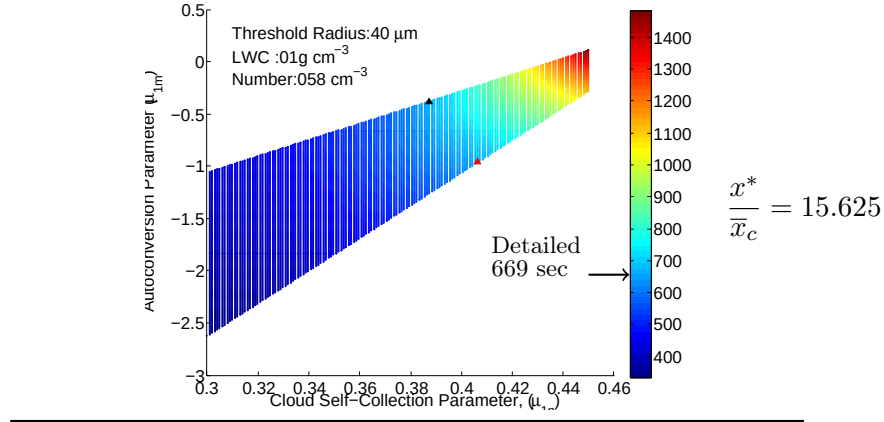


Figure 5.13: The black, and red, marks locate the stochastic parameters: (0.38742, -0.38000) and (0.40629, -0.96000), respectively. When these parameters drive the bulk model, 50% of the cloud mixing ratio converts to rain mixing ratio in 665 and 663 seconds, respectively.

time step of 50.4 and 40.0 μm , respectively.

The iso-lines on Figure 5.13 are similar to the those Figure 5.5 in that they show a comparable effect by the two competing parameters. However, these isolines have a stronger curvature. When stochastic parameters give a mean rain radius of 40.0 μm at the first time step, the 50% conversion time is sensitive to the auto conversion parameter, but not the cloud self-collection parameter. Conversely, when stochastic parameters produce the largest possible mean rain radius at the first time step, the 50% conversion time is more sensitive to the cloud self-collection parameter than the auto conversion parameter.

The two pairs of stochastic parameters marked on Figure 5.13 are detailed in Table 5.4. As expected, a larger mean rain radius at the first time step produces a larger mean rain radius at the time of 50% conversion, which is seen when comparing the first two rows and when comparing the last two rows. The difference between the mean rain radius at the time of 50% conversion is less pronounced than in the polluted case, but the conversion time is also short, nearly a third of the time required by the polluted parameterized case.

Table 5.4: Parameterized Clean: Stochastic Parameters

(μ_{1n}, μ_{1m})	$q_c = q_r$	$t = 1 \text{ second}$	$\overline{x}_r = x^*$
	seconds	$\overline{x}_r \text{ (}\mu\text{m)}$	$\overline{x}_r \text{ (}\mu\text{m)}$
			seconds
$(0.38742, -0.38000)$	665	93.9	40.0
$(0.40629, -0.96000)$	663	129.7	50.4

Table 5.4: The same as Table 5.1 except the stochastic parameters are found in the contour map in Figure 5.13.

The two stochastic curves (Stoch A and Stoch B) in Figure 5.14 correspond to the two rows in Table 5.4. They also correspond to the red and black triangles in Figure 5.13, respectively. These two stochastic evolution curves are compared to a detailed curve produced by Bott's LFM detailed method. We see a stark difference in the evolution curves between the deterministic one and the stochastic ones. Similarly to the clean hydrodynamic case, the stochastic ones begin with a strong conversion rate followed by a weaker rate until the conversion accelerates rapidly at approximately 2000 seconds and 2400 seconds for Stoch A and Stoch B, respectively. Again, a more complex model such as was mentioned in Section 5.1.1 may be useful to determine if this loss of cloud mass corresponds with cloud termination.

Parameters of the Stochastic curves in Figure 5.14:

Stoch A : $\mu_{1n} = 0.40629$, and $\mu_{1m} = -0.96000$

Stoch B : $\mu_{1n} = 0.38742$, and $\mu_{1m} = -0.38000$

As in the polluted case, the mean rain droplet radius, both at the first time step and at the 50% conversion time, are plotted as functions of the cloud self-collection and auto conversion parameters in Figure 5.7. The contour of the mean rain droplet radius at the 50% conversion time is plotted in the graph on the left. We again see a strong dependence on the cloud self-collection parameter and very little dependence on the auto conversion parameter. This pattern is disrupted when the conversion time is delayed. However, the disruption is less pronounced than when the initial cloud radius is a smaller fraction of the threshold radius.

Figure 5.15a again shows a smooth transition from the mean rain radius at the first time step of 40 to 50 μm as was present in the polluted case.

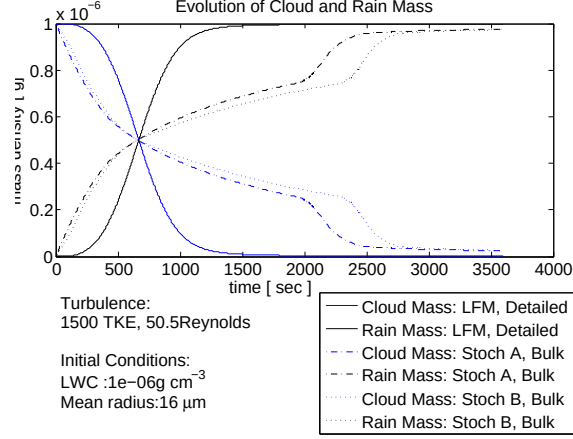
Figure 5.14: Parameterized, Clean: Evolution Curves

Figure 5.14: Bulk evolution of mass conversion comparing the stochastic model benchmarked against a detailed method for a clean cloud with an initial mean droplet radius of 16 μm.

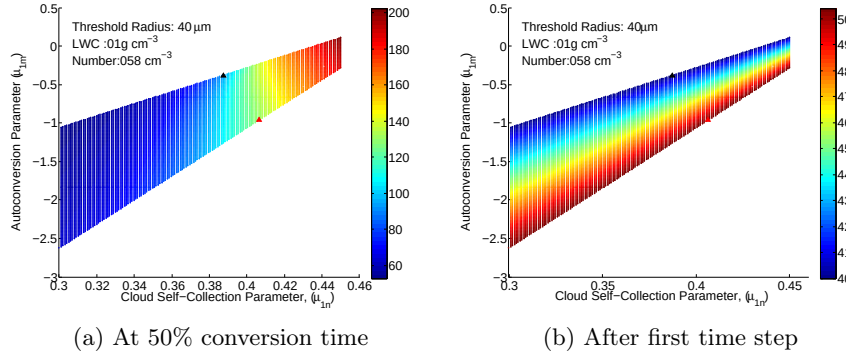
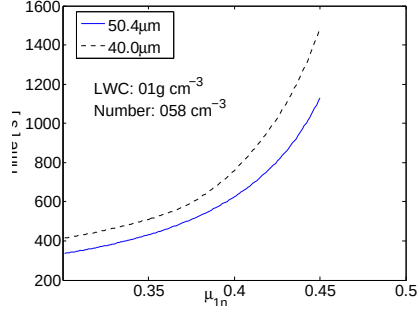
Figure 5.15: Parameterized, Clean: Mean Rain Radius

Figure 5.15: The mean rain droplet radius at the time of 50% mass conversion is graphed in (a). The mean rain droplet radius at the first time step as a function of the cloud-self-collection and auto conversion parameters for a polluted cloud with an initial mean droplet radius of 16 μm is graphed in (b).

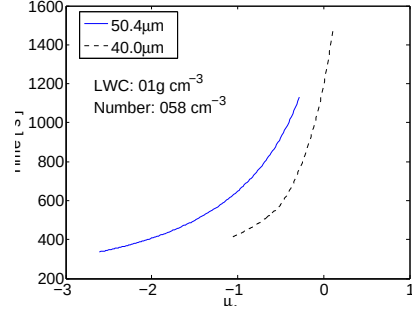
The two of the edges of the triangle in Figures 5.13 and 5.15 are detailed in Figure 5.16. Figures 5.16a and 5.16b show the 50% conversion time as a function of each parameter. Figures 5.16c and 5.16d show the mean rain radius as a function of each parameter. Note that the upper edge of the

triangle has a higher lower bound of values of μ_{1m} than the lower edge. Therefore, the black curves on the right two plots do not have the same domain as the blue curves.

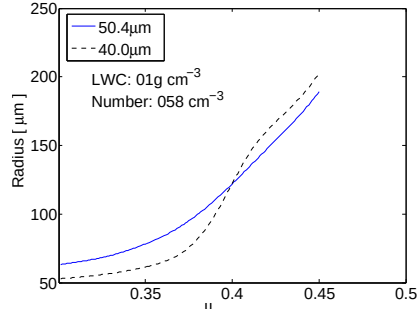
Figure 5.16: Parameterized, Clean: Edge Plots



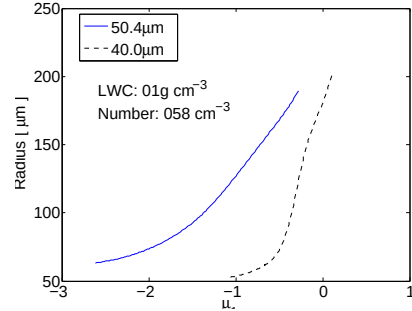
(a) 50% conversion time over cloud self-collection parameter



(b) 50% conversion time over cloud self-collection parameter



(c) 50% conversion time over cloud self-collection parameter



(d) 50% conversion time over cloud self-collection parameter

Figure 5.16: Same as Figure 5.4 except the bulk stochastic model uses Franklin's (2007) parameterized turbulent kernel.

The slices along the edges of the contour triangles in both the clean and the polluted parameterized cases have strong similarities with the respective hydrodynamic cases.

5.3 Tabulated Turbulent Kernel

The turbulent kernel used in this subsection is by Pinsky Khain (2008) and is further described in Section 2.2.3. The simulations performed in this section used turbulent values that are at the upper bounds of the valid range for their parameterization ($1000 \text{ cm}^2\text{s}^{-3}$ and 50.5 Reynolds number). The turbulent enhancement factor for this collision rate is shown in the right contour plot of Figure 2.2, and can be characterized as having a stronger typical large scale air flow in the cloud, but a weaker viscous dissipation rate. The contour graph indicates that the strongest enhancement occurs for smaller droplets collected by the larger cloud droplets ($15\text{--}21 \mu\text{m}$). Turbulent phenomena dependent on the droplets' different velocity or inertia, such as fluid acceleration and shear, are the dominant forces effecting droplets with these characteristics (Wang et al. 1998, Pruppacher & Klett 1997).

Figure 5.17: Tabulated, Polluted: Contour, 50% conversion

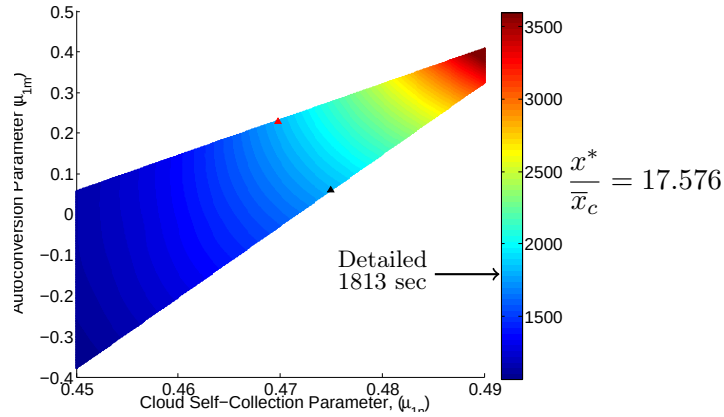


Figure 5.17: The black, and red, marks locate the stochastic parameters: (0.47496, 0.06000) and (0.46975, 0.22900), respectively. When these parameters drive the bulk model, 50% of the cloud mixing ratio converts to rain mixing ratio in 1813, and 1811, seconds, respectively.

5.3.1 Polluted Cloud: $239 \text{ cm}^3\text{s}^{-1}$

Figure 5.17 shows the contours of the 50% conversion time for the stochastic bulk method as a function of the parameters μ_{1n} and μ_{1m} . The 50% conversion time for the detailed method (1813 sec) is shown on the colour bar. Similar to the parameterized turbulent polluted case, the value for the cloud self-collection parameter is limited to those values that are close to the maximum 0.5 which reduces the strength of the auto conversion process in the stochastic bulk model. Here, the red and black marks are used in a similar manner as they are in the parameterized turbulent cases.

The iso-lines on Figure 5.17 for this polluted case is marked different than for the previous two polluted cases. Here the iso-lines show that the 50% conversion time has a comparable dependence on both parameters. This is similar to the 50% contour plots for both the hydrodynamic and parameterized turbulent clean cloud cases. However by visual inspection, on the region of the contour plot in Figure 5.17 near the detailed solution, the curvature of the contour lines is less than they are for the two cases in Figures 5.5 and 5.13, and we don't see one parameter dominate the other edges of the triangle as we did in the parameterized clean case.

Table 5.5: Tabulated, Polluted: Stochastic Parameters

(μ_{1n}, μ_{1m})	$q_c = q_r$		$t = 1 \text{ second}$
	seconds	$\bar{x}_r \text{ } (\mu\text{m})$	$\bar{x}_r \text{ } (\mu\text{m})$
(0.46975, 0.22900)	1811	155.8	26.2
(0.47496, 0.06000)	1813	160.3	32.8

Table 5.5: The same as Table 5.1 except the stochastic parameters are found in the contour map in Figure 5.17.

The two pairs of stochastic parameters marked by triangles (one red and one black) on Figure 5.17 are detailed in Table 5.5. As with the previous polluted cases, a larger mean rain radius at the first time step produces a larger mean rain radius at the time of 50% conversion, which is seen when comparing the two rows. The difference between the mean rain radius at the time of 50% conversion is again less pronounced than in the hydrodynamic case, but it should be noted that the values for both stochastic parameters in the turbulent cases lead to stronger cloud self-collection and auto conversion

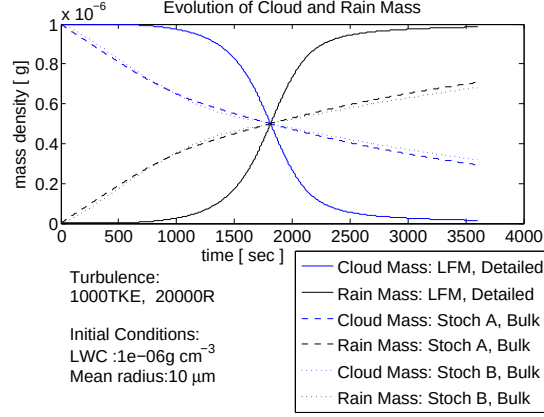
Figure 5.18: Tabulated, Polluted: Evolution Curves

Figure 5.18: Bulk evolution of mass conversion comparing the stochastic model benchmarked against a detailed method for a polluted cloud with an initial mean droplet radius of $10 \mu\text{m}$.

processes in the bulk model than the values for the hydrodynamic case.

The two stochastic curves (Stoch A and Stoch B) in Figure 5.18 correspond to the two rows in Table 5.5, respectively. They also correspond to the black and red triangles in Figure 5.17, with a mean rain radius at the first time step of 40 and 50.4, respectively. Again, we see a stark difference in the evolution curves between the deterministic ones and the stochastic ones. As with the other two polluted cases, the stochastic ones have a near linear conversion rate. However, here we don't see a final cloud mass loss as we did in previous cases, but we do see a change in the linear rate near 1000 seconds.

The mean rain droplet radius, both at the first time step and at the 50% conversion time, are plotted as functions of the cloud self-collection and auto conversion parameters in Figure 5.19. The contour of the mean rain droplet radius at the 50% conversion time is plotted in the graph on the left. In the region on this contour plot near the 50% conversion time, there is very little variation in mean rain drop radius. This is in contrast to the previous four cases.

Parameters of the Stochastic curves in Figure 5.18

Stoch A : $\mu_{1n} = 0.46975$, and $\mu_{1m} = 0.22900$

Stoch B : $\mu_{1n} = 0.47496$, and $\mu_{1m} = 0.06000$

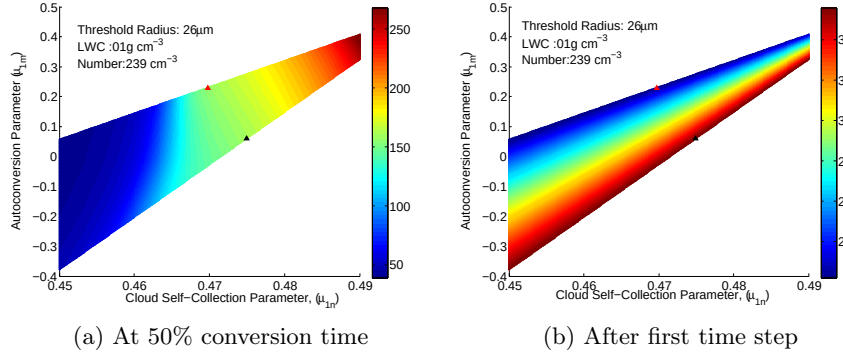
Figure 5.19: Tabulated, Polluted: Mean Rain Radius

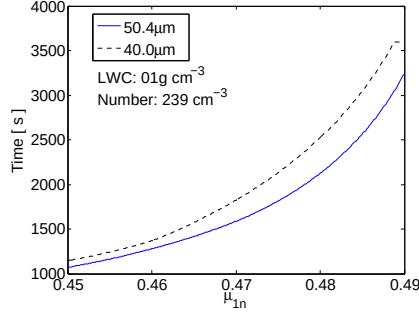
Figure 5.19: The mean rain droplet radius at the time of 50% mass conversion is graphed in (a). The mean rain droplet radius at the first time step as a function of the cloud-self-collection and auto conversion parameters for a polluted cloud with an initial mean droplet radius of $10\mu\text{m}$ is graphed in (b).

The mean rain radius at the first time step (Figure 5.19b) show the same pattern as both the clean and polluted hydrodynamic and parameterized turbulent cases.

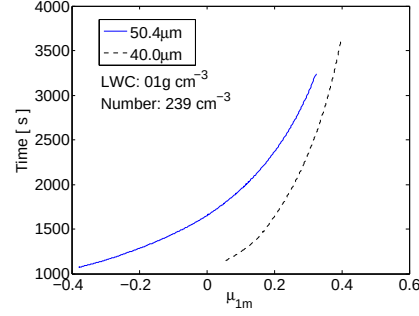
The two of the sloped edges (upper and lower) of the quadrilateral in Figures 5.17 and 5.19 are detailed in Figure 5.20. These four graphs plot the same information as what were shown in each hydrodynamic case. They give very similar results to what were shown for both of the previous polluted cases in all four plots except that the ranges of parameter values are obviously more similar to the ranges for the parameterized case than the hydrodynamic one, except that in this tabulated turbulent case the mean rain radii at the 50% conversion time does not reach a maximum value before the end of the simulation.

5.3.2 Clean Cloud: $58.3 \text{ cm}^3\text{s}^{-1}$

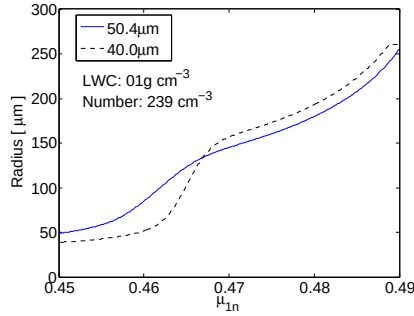
Figure 5.21 shows the contours of the 50% conversion time for the stochastic bulk method as a function of the parameters μ_{1n} and μ_{1m} . The 50% conversion time for the detailed method (699 sec) is shown on the colour bar. Unlike the simulations for the polluted tabulated turbulent cloud, the clean cloud

Figure 5.20: Tabulated, Polluted: Edge Plots

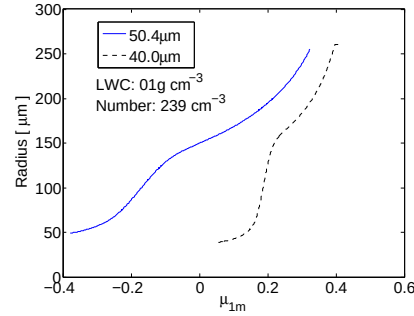
(a) 50% conversion time over cloud self-collection parameter



(b) 50% conversion time over cloud self-collection parameter



(c) 50% conversion time over cloud self-collection parameter



(d) 50% conversion time over cloud self-collection parameter

Figure 5.20: Same as Figure 5.4 except the bulk stochastic model uses Pinsky and Khain's (2008) tabulated turbulent kernel.

has an initial mean cloud radius that is closer to the threshold radius ($40\mu\text{m}$). Additionally, unlike the previous clean cloud cases, the minimum value for μ_{1n} extends to 0.45, 5% of the range in the clean hydrodynamic case. Again, the red and black triangles locate stochastic parameter pairs that approximate the 50% conversion time for the detailed method given a mean rain radius at the first time step of 50.4 and 40.0 μm , respectively.

The contour lines on Figure 5.21 appear to have a strong linear quality near the region containing the 50% conversion time produced by the detailed method. Linear approximations to these contour lines have a different sign to the slope, than approximations to the contour lines on any of the other

Figure 5.21: Tabulated Clean: Contour, 50% conversion

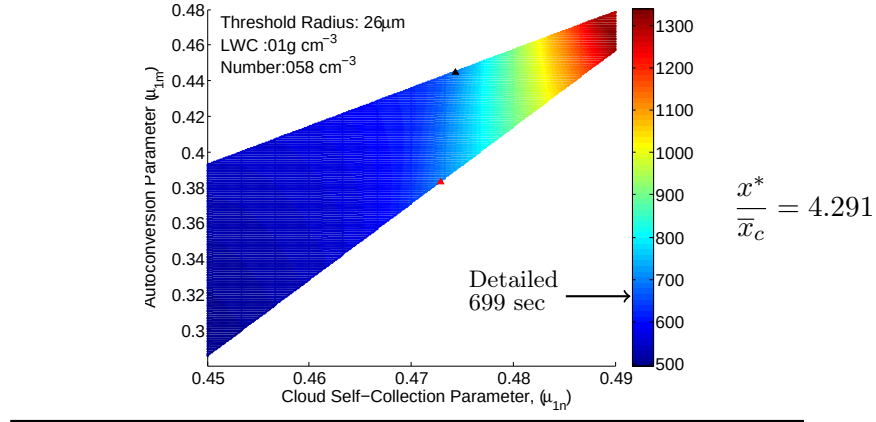


Figure 5.21: The red, and black, marks locate stochastic parameters: (0.47285, 0.38353) and (0.47437, 0.44490), respectively. When these parameters drive the bulk model, 50% of the cloud mixing ratio converts to rain mixing ratio in 698, and 699, seconds, respectively.

50% conversion contour graphs. Using the red mark as a reference, and remembering that at the edge near the red mark the largest cloud droplets dominate auto conversion, a stronger cloud self-collection and a weaker auto conversion produce a similar 50% conversion time. While in the two other clean cases, weaker cloud self-collection and weaker auto conversion processes produced a similar 50% conversion time.

The two pairs of stochastic parameters marked on Figure 5.21 are detailed in Table 5.6. As expected, a larger mean rain radius at the first time step produces a larger mean rain radius at the time of 50% conversion, which is seen when comparing the two rows. Again, the difference between the mean rain radius at the time of 50% conversion is less pronounced than in the polluted case, but the conversion time is also short, nearly a third of the time required by the polluted parameterized case.

Parameters of the Stochastic curves in Figure 5.22:

Stoch A : $\mu_{1n} = 0.47285$, and $\mu_{1m} = 0.38353$

Stoch B : $\mu_{1n} = 0.47437$, and $\mu_{1m} = 0.47437$

Table 5.6: Tabulated Clean: Stochastic Parameters

(μ_{1n}, μ_{1m})	$q_c = q_r$	$t = 1 \text{ second}$	
	seconds	$\bar{x}_r (\mu\text{m})$	$\bar{x}_r (\mu\text{m})$
(0.47285, 0.38353)	697	143.6	26.0
(0.47437, 0.44490)	699	114.0	26.0

Table 5.6: The same as Table 5.1 except the stochastic parameters are found in the contour map in Figure 5.21.

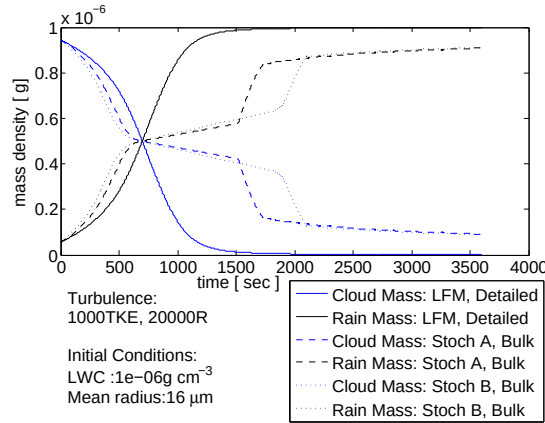
Figure 5.22: Tabulated, Clean: Evolution Curves

Figure 5.22: Bulk evolution of mass conversion comparing the stochastic model benchmarked against a detailed method for a clean cloud with an initial mean droplet radius of 16 μm.

The two stochastic curves (Stoch A and Stoch B) in Figure 5.22 correspond to the two rows in Table 5.6. They also correspond to the red and black triangles in Figure 5.21, respectively. These two stochastic evolution curves are compared to a detailed curve produced by Bott's LFM detailed method. Prior to the 50% conversion time, the mass conversion is fairly similar. However, it becomes starkly different after this threshold. Similarly to the clean hydrodynamic case, the stochastic ones begin with a strong conversion rate followed by a weaker rate until the conversion accelerates rapidly at approximately 1500 seconds and 1900 seconds for Stoch A and Stoch B, respectively. Again, a more complex model such as was mentioned in Section 5.1.1 may be useful to determine if this loss of cloud mass corresponds with cloud

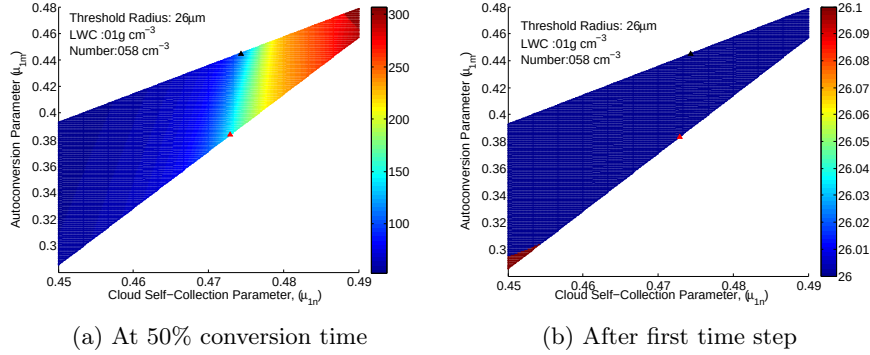
Figure 5.23: Tabulated, Clean: Mean Rain Radius

Figure 5.23: The mean rain droplet radius at the time of 50% mass conversion is graphed in (a). The mean rain droplet radius at the first time step as a function of the cloud-self-collection and auto conversion parameters for a polluted cloud with an initial mean droplet radius of $16\mu\text{m}$ is graphed in (b).

termination.

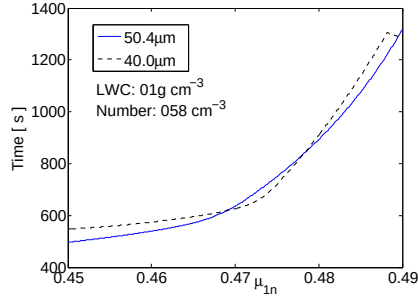
As in the polluted case, the mean rain droplet radius, both at the first time step and at the 50% conversion time, are plotted as functions of the cloud self-collection and auto conversion parameters in Figure 5.23. The contour of the mean rain droplet radius at the 50% conversion time is plotted in the graph on the left. We again see a strong dependence on the cloud self-collection parameter and very little dependence on the auto conversion parameter. This pattern is uniform throughout the region displayed.

Figure 5.23b shows uniformity of the mean rain radius at the first time step at $26.0\mu\text{m}$. This may be due to the narrow range of parameters near the strong cloud self-collection/ weak auto conversion limit of $(\mu_{1n}, \mu_{1m}) = (0.5, 0.5)$ and the closeness of the initial mean cloud radius of $16\mu\text{m}$ and the threshold radius of $26\mu\text{m}$. However, the reason is unknown and more research into the stochastic parameter space is needed.

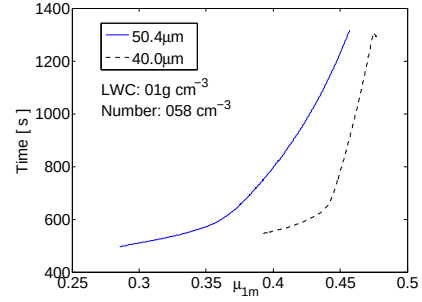
The two of the sloped edges (upper and lower) of the quadrilateral in Figures 5.21 and 5.23 are detailed in Figure 5.24. These four graphs plot the same information as what were shown in each hydrodynamic case. They give very similar results to what were shown for the previous clean cases in all four plots except where the parameter values are near 0.5. There the curves representing the mean rain radius and time of 50% conversion display an

erratic behaviour. The cause is unknown, but this could be related to a non-trivial amount of liquid water content initializing as rain because this is one condition that is not present in the other five simulations. More research into the stochastic parameter space may explain this phenomena.

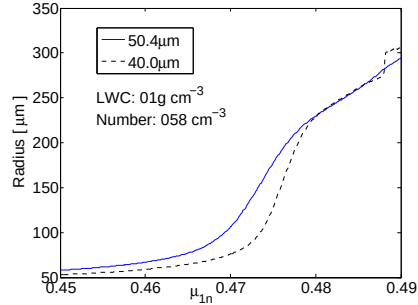
Figure 5.24: Tabulated, Clean: Edge Plots



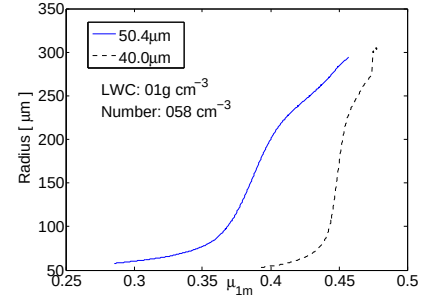
(a) 50% conversion time over cloud self-collection parameter



(b) 50% conversion time over cloud self-collection parameter



(c) 50% conversion time over cloud self-collection parameter



(d) 50% conversion time over cloud self-collection parameter

Figure 5.24: Same as Figure 5.4 except the bulk stochastic model uses Pinsky and Khain's (2008) tabulated turbulent kernel.

5.4 Summary

Three types of contour maps are each presenting a result of the stochastic bulk model of collision and coalescence: 50% conversion time, mean rain radius at the time of the first time step, and mean rain radius at the time of 50% conversion time. Results for each of these three types are presented for three kernels and two initial droplet concentrations. The three kernels are (i) a hydro dynamic one, (ii) a parameterized turbulent one that predominantly enhances coupling, and (iii) a tabulated one that most strongly enhances turbulent phenomena associated with fluid velocity differentials. The two initial droplet concentrations are 239 cm^{-3} and 58.3 cm^{-3} which correspond with an initial mean droplet radii of $10\mu\text{m}$ and $16\mu\text{m}$. An existing turbulent bulk model was not presented because the purpose of this chapter is to extend to applicability of the stochastic bulk model, not to further validate it with a turbulent kernel. The further validation with a turbulent kernel would best be done after a through exploration of the stochastic parameter space. Further exploration of acquiring parameter values beyond the initial exploration in Chapter 6 would make use of other existing bulk models. Such work is beyond the scope of this dissertation which is limited to derivation, validation, and extension.

The set of six contour maps showing the 50% conversion time gives information on the relationship between ratio of the threshold mass to the mean cloud mass ($x^*/\bar{x}_c$) and the relative strength of auto conversion to cloud self-collection given a constant 50% conversion time. The contour lines for the detailed method are present in each of Figures 5.1, 5.5, 5.9, 5.13, 5.17, and 5.21. A qualitative linear approximation can be visualized for each graph:

Characteristic Slope $\approx 0, -1, 0, -1, -1$, and $+$, respectively.

Associated with each figure is the relationship between the threshold mass and the initial mean droplet mass:

$$\frac{x^*}{\bar{x}_c} \approx 64, 15.625, 64, 15.625, 17.576, \text{ and } 4.29, \text{ respectively.}$$

For a small initial mean droplet mass ($x^*/\bar{x}_c=64$) the cloud self-collection parameter has small effect relative to the auto conversion parameter for a constant 50% conversion time. For a larger mean cloud mass and smaller ratio (≈ 15) a constant 50% conversation time is achieved by decreasing the

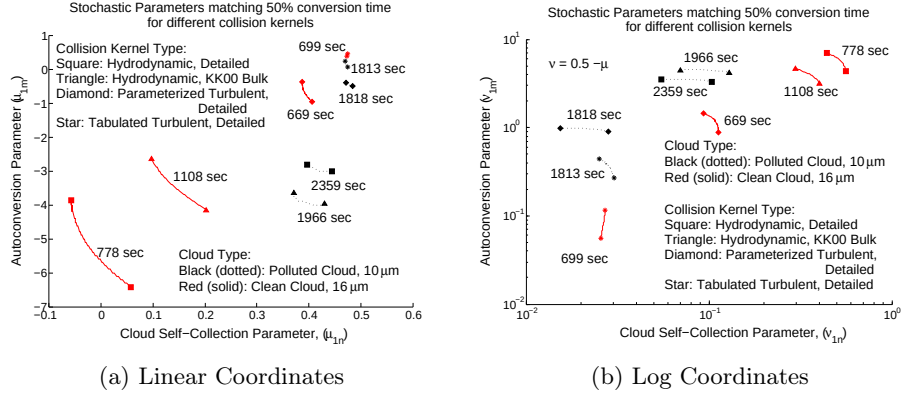
Figure 5.25: Stochastic Parameters, Six Simulations

Figure 5.25: Each graph plots eight loci of stochastic parameter pairs that match the 50% conversion time for different collision kernels. The six simulations span three kernels: a hydrodynamic one, and two turbulent kernels. For each kernel there is a simulation for a clean cloud and for a polluted cloud. Both graphs plot the contours lines from Figures 5.1, 5.5 corresponding to a detailed simulation and to the KK00 bulk model, and Figures 5.9, 5.13, 5.17 5.21 corresponding to a detailed simulation. Linear scales are used in (a). We apply $\nu = 0.5 - \mu$ to the stochastic parameters and plot on a log scale in (b).

strength of both the cloud self-collection process and the auto conversion process. As the mean cloud mass approaches the threshold mass ($x^*/\bar{x}_c \approx 4$), a constant 50% conversation time is achieved by decreasing the strength of auto conversion and increasing the strength of cloud self-collection. These results are the same for the hydrodynamic and parameterized turbulent kernels at both initial droplet concentrations.

A possible explanation or why the evolution curve generated by the stochastic bulk model are so different from the one generated by the detailed method could be because the stochastic parameters were taken as averages over the entire simulation instead of averaging over separate time intervals that correspond to when different collision and coalescence phenomena are dominate. Specifically, more research could be done which uses stochastic parameters when cloud self-collection is dominant, when accretion is dominant, and when rain self-collection is dominant. Auto conversion has been shown to not be a dominant process for a specific turbulent collision kernel (Franklin 2008).

The contour graphs showing rain radius at the time of 50% conversion show

a similar concave structure in the smaller radius region and a linear structure in the larger radius region. This relationship hold for the hydrodynamic and parameterized turbulent kernels, which occurs when the ratio is larger $x^*/\bar{x}_c$. However, the tabulated kernel shows a markedly different structure even for a similar ratios: 15.625 and 17.576, in the region of smaller rain radius. Compare the left graphs in Figures 5.15 and 5.19. For the extreme ratio $x^*/\bar{x}_c \approx 4$, the region of the smaller rain radius adopts the linear structure found in the region of the larger rain radius.

Except for the case of $x^*/\bar{x}_c \approx 4$, there is a smooth transition from the minimal initial mean rain radius to the maximal initial mean rain radius. This is consistent for three kernels and both initial droplet concentrations.

The two graphs in Figure 5.25 plot the stochastic parameters pairs for six different kernels that yield a 50% conversion time that matches conversion times from reference simulations (detailed and KK00). Figure 5.25a uses the linear scales of the figures in Sections 5.1, 5.2, and 5.3. Figure 5.25b applies $\nu = 0.5 - \mu$ to the stochastic parameters and plots on a log scale for the purpose of making clearer the contours near $(\mu_{1n}, \mu_{1m}) = (0.5, 0.5)$. The purpose of these two graphs and the reason for ending this chapter with them is to emphasize the generality of the stochastic bulk parameterization, given in Equation 4.1, and its applicability to a variety of cloud types and kernels by simply choosing appropriate stochastic parameters with the relevant collision kernel.

6 | Parameters

In Chapter 5 we extended the stochastic bulk model to include turbulent collision kernels and graphically acquired values for two of the parameters in that model. We selected a metric based on conversion of cloud mass to rain mass. Specifically, parameters were implicitly acquired based on the time required for the stochastic model to convert 50% of cloud mass to rain mass. The parameters which produced a 50% conversion time that matched the 50% conversion time given by a detailed method, Linear Flux Method (LFM) (Bott 1998) were chosen. A collection of 3-D contour maps were used to make this comparison. In this chapter we take a different approach to determining the stochastic parameters by acquiring the values explicitly from the detailed evolution of the droplet spectrum.

6.1 Acquired from 0th Order Deterministic Method

The same detailed method (Bott's Linear Flux Method) is used to explicitly acquire values for the stochastic parameters. Bott's Linear Flux Method is a 0th Order Deterministic Method. At each time step, the state space mean of the four partial moments (N_c , N_r , q_c , and q_r) are calculated. For each point in the number and mass distributions, the fluctuations from the mean are calculated:

$$\begin{aligned}\delta_c &= N_c - \overline{N}_c, & \delta_r &= N_r - \overline{N}_r \\ \gamma_c &= q_c - \overline{q}_c, & \gamma_r &= q_r - \overline{q}_r\end{aligned}$$

These fluctuations over the cloud portion ($0 \leq x \leq x^*$), or the rain portion ($x \geq x^*$) of the droplet size spectrum are summed. The summation occurs

in the form of a double integral as shown in Equations 3.15, 3.22, 3.30, and 3.37.

Figure 6.1: Hydrodynamic, LFM

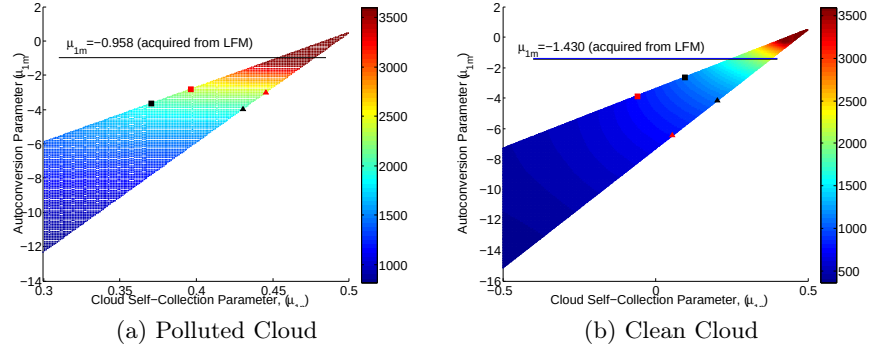


Figure 6.1: Same as Figures 5.1 and 5.5 except the value for μ_{1m} from the detailed evolution of the droplet spectrum is included in each contour plot. Whether the acquired parameter (μ_{1m}) falls within the region of validity depends on the value of μ_{1n} that would be chosen.

These instantaneous, aggregate fluctuations of the four evolving quantities are recorded at each time step. The means (μ_*) of the aggregate fluctuations for each evolving quantities are the stochastic parameters, and variances (ω_*) are dismissed by order of magnitude arguments in Sections 3.3.1.

The Linear Flux Method evolves mass only (Bott 1998). Number concentration is calculated using the mass (on the discretized spectrum) and the mass density at the bin's centre point. The LFM assumes a constant intra-bin distribution of mass density which produces large errors when the droplet size densities changes rapidly (Wang et al. 2007). Therefore the fluctuations associated with number concentration are not calculated from the evolution of the droplet size distribution by the LFM. Only values for the stochastic parameter μ_{1m} are collected.

The parameter reduction (from five to three) in Section 3.3.4 showed that $\mu_{1n} = -\mu_{2n}$ and $\mu_{1m} = -\mu_{2m}$. Both mass parameters are calculated independently. For each of the six simulations, these two parameter values are shown on the respective contour graphs in Figures 6.1, 6.2, and 6.3. Where the two values are indistinguishable at three decimal places, only one value is provided and graphed.

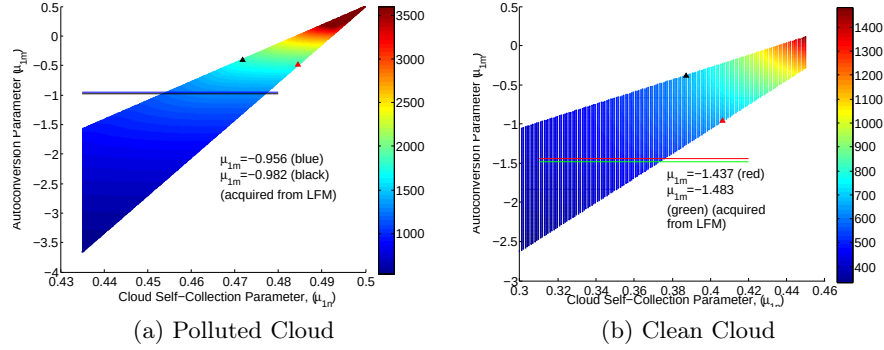
Figure 6.2: Parameterized Turbulent, LFM

Figure 6.2: Same as Figures 5.9 and 5.13 except the values for μ_{1m} from the detailed evolution of the droplet spectrum is included in each contour plot.

6.1.1 Hydrodynamic Kernel

Simulations for the polluted, and the clean, cloud using the hydrodynamic kernel produced the same stochastic parameters for μ_{1m} and μ_{2m} (to three decimal places). The value for these parameters evolve the droplet distribution in the stochastic model much slower than the detailed method by noting that the horizontal lines for μ_{1m} in both graphs in Figure 6.1 indicate a slower 50% conversion time than any of the red or black marks.

The 50% conversion times can be estimated using the colour bars to the right of each of the contour plots in this section. For exact values refer to Section 5.1. The mean rain radii are not present on the graph and are presented in the text for convenience. The triangular markers correspond to mean rain radii, at the first time step, of 50.4, and the diamond and squares to 40.0 μm for both plots. For the polluted contour plot (Figure 6.1a), the mean rain radii at the 50% conversion time associated with the detailed (red) markers are 226.2 and 176.3 μm for the triangular and diamond, respectively. For the clean cloud (Figure 6.1b), the detailed (red) markers correspond with 138.8 and 108.6 μm for the triangular and square, respectively.

6.1.2 Parameterized Turbulent Kernel

The contour plots of the parameterized turbulent kernel have the same shape as the hydrodynamic ones: $x^*/\bar{x}_c = 64$ and ≈ 15 , for Figures 6.2a and 6.2b,

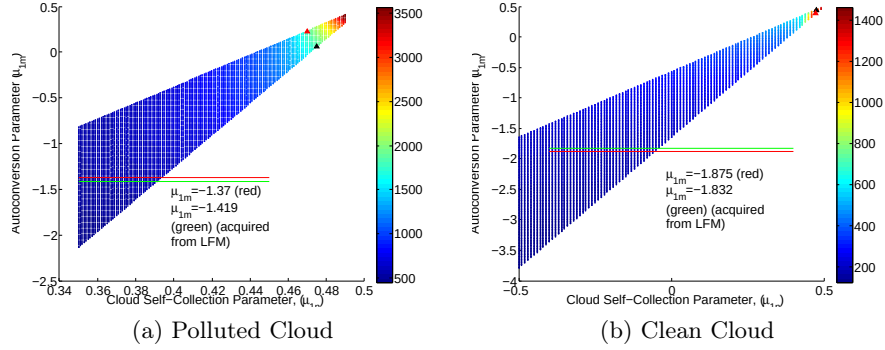
Figure 6.3: Tabulated Turbulent, LFM

Figure 6.3: Same as Figures 5.17 and 5.21 except the value for μ_{1m} from the detailed evolution of the droplet spectrum is included in each contour plot.

respectively. Despite the similar geometries, the hydrodynamic and turbulent kernels generate different parameter values relative to the markers in the contour plots. The marks identify parameter values that produce the same 50% conversion time as the detailed evolution. In contrast to the parameter values for the hydrodynamic kernel, the ones for the parameterized turbulent kernel unnaturally accelerate the evolution of the droplet size distribution. As the acceleration becomes more pronounced, the difference between μ_{1m} and μ_{1n} become discernible.

The 50% conversion times can be estimated using the colour bars to the right of each graph. For exact values refer to Section 5.2. As usual, the red and black markers correspond to initial mean rain radii of 50.4 and 40.0 μm , respectively for both the polluted and clean plots. For the polluted contour plot (Figure 6.2a), the 50% conversion time mean rain radii associated with the red and black markers are 210.5 and 124.3 μm , respectively. For the clean cloud (Figure 6.2b), the red and black markers correspond with 129.7 and 93.9 μm , respectively.

6.1.3 Tabulated Turbulent Kernel

Because of the much smaller ratio of $x^*/\bar{x}_c$ (4.29 vs. 17.576 and 64), the shape of the clean cloud contour plot of the tabulated turbulent kernel has a different orientation than the corresponding ones generated by the other two kernels. This may not be readily apparent because the scales of the axes

between contour plots are not consistent. The ratio of the mass threshold and the mean initial radius is ≈ 17.576 and 4.29 for the polluted and clean clouds, respectively. Despite the contour plot for the polluted cloud (Figure 6.3a) having a similar shape as the clean cloud plots Figures 6.1 and 6.2, the stochastic parameters acquired from detailed simulations, relative to stochastic parameters giving the same 50% conversion time as evolution of the droplet size spectrum by detailed simulations, vary considerably by kernel. Detail simulations by the tabulated kernel are much more accelerated than the ones by the parameterized kernel, which is more accelerated than the simulations by the hydrodynamic kernel.

It appears that the parameter acquisition by detailed simulations are sensitive to the choice of kernel. It appears that Bott's Linear Flux Method is prone to sufficient error, by virtue of the intra-bin distribution assumptions, that it does not produce values of the parameters in the stochastic bulk model that are reasonably close to those values necessary to approximate the 50% conversion rate of the detailed method.

6.2 Proposed Acquisition Method: Modified Detailed

Two parameter value acquisition methods have been presented: implicitly determining values using a 3-D contour plot (graphical) in Chapter 5, and explicitly calculating values using a detailed evolution of the droplet size distribution in Section 6.1. The detailed method of parameter value acquisition can be modified.

For each of the three parameters in Equation 4.1, values have been acquired during the times when different collision processes were dominant. Values for μ_{1n} , μ_{1m} , and μ_{4np} were acquired when cloud self-collection was dominant and when accretion was dominant. For all four kernels auto conversion was never dominant. This is consistent with results obtained by Franklin (2008).

An example of the differences in parameter values can be seen in Figure 6.4. It shows the evolution of the mean value of the rain self-collection parameter, as well as showing when accretion becomes more dominant than cloud self-collection. Cloud self-collection is dominant for the first 297 seconds. During this time the mean value of the rain self-collection parameter is 0.3625. This

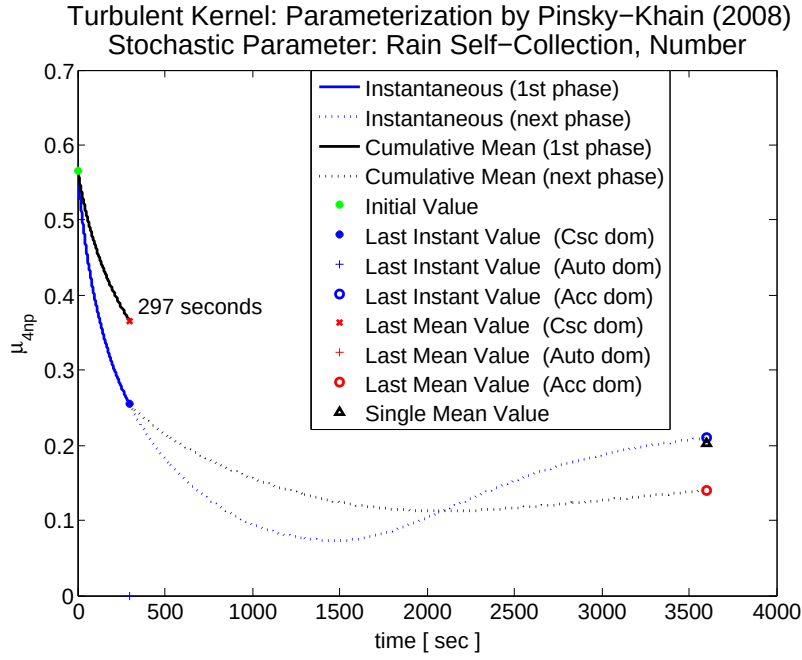
Figure 6.4: Double Value: Rain Self-Collection

Figure 6.4: Rain self-collection parameter μ_{4np} during a simulation using the tabulated collision kernel. Initial conditions: 1 g cm^{-3} and an initial mean cloud radius of $14 \text{ }\mu\text{m}$.

value inhibits the rain self collection term as 0.75 will completely shut down this process in the stochastic bulk model (i.e. consistency of number requires $(0.75 - \mu_{4nc}) \geq 0$ in the second equation in Equation 4.1) and 0.2029 is the mean value over the entire one hour simulation. The mean value over the remaining 3303 seconds is 0.1885 which is only a slight acceleration over the single parameter value.

Other possible modifications to the parameter value collection method using a detailed simulation is to vary the time step or to use a metric different than the 50% mass conversion time. Although reducing the time step for the LFM doesn't improve the accuracy of the evolution of the droplet spectrum, that is not an indication that the parameter collection values won't be effected by a shorter time step. Targeting a different conversion threshold than 50%, such as 20% or 80%, may give different values than the ones produced by the 50% threshold.

In addition to different approaches using the LFM, a different detailed method, such as one that assumes a linear (Wang et al. 2007) or quadratic (rather than constant) intra-bin distribution of mass density, could give appropriate stochastic parameter values. Some partial results towards a quadratic intra-bin distribution algorithm are presented in Section 7.2.1, and some partial results towards a modification of the Gillespie-Seeßelberg Algorithm are also presented in Section 7.2.2.

6.3 Summary

The graphical method of selecting stochastic parameter values is more flexible than the current method of acquiring values using a detailed algorithm that assumes a constant intra-bin distribution of mass density. The graphical method also produces values that give reasonable evolution curves compared to the detailed parameter values. Different metrics, than the 50% mass conversion threshold, could be used to select parameter values. Also, other detailed methods could be used to select parameter values: deterministic ones that assume a linear or quadratic intra-bin distribution of mass density, or a stochastic simulation.

7 | Discussion

In this Chapter, we present theoretical and simulation results from the stochastic bulk model. We also comment on non-graphical methods of acquiring stochastic parameter values, such as a new detailed bin-pair interaction method and for a stochastic simulation method. The partial results are detailed in Chapter B and guide the direction that future research can take with regards to acquiring stochastic parameters from non-graphical methods.

7.1 Stochastic Bulk Model

A novel methodology in the derivations of the stochastic bulk model of collision and coalescence produced several unprecedented and desirable model features. The novel methodology is to represent state space uncertainty as a mean and a set of point-wise fluctuations, use physical characteristics of the phenomena to reshape the state space fluctuations into fluctuations of a stochastic process, and use established methods to reduce the variations in the stochastic processes.

The current application of this methodology involved a numerical solution of moments of the kinetic collection equation (KCE). Solutions of moments of the KCE, for all but the simplest of collision kernels, introduce a closure problem. The first moment solution has a second moment term, and the second moment term depends on a third moment term, etc. The closure problem is handled nicely by representing the evolution of these higher moment terms as stochastic processes, which is to the author's knowledge, a new way to address the closure of the KCE.

Using a mean and a set of point-wise fluctuations eliminates the need to assume a probability distribution, and allows each of the four equations in the coupled set to be derived independently. This is in contrast to all other bulk models that would derive some terms and make remaining terms to be functions of the derived terms. By independently deriving all equations, the conservation of mass is intrinsic to the derivations rather than being applied externally. No other bulk models that the author is aware of demonstrate an intrinsic conservation of mass. An intrinsic conservation of mass is desirable because it validates the physical authenticity of the derivations and the model.

Each of the four equations contains an auto conversion term. Auto conversion is the only collision and coalescence process that occurs in all four equations. It is the process most effected by turbulence, and the one that is least understood. By independently deriving all four auto conversion terms, we are able to control the auto conversion parameter in an unprecedented manner. No other known bulk models has an auto conversion parameter that is entirely based in the physics of the problem, and that is also controlled in a finely tuneable manner.

We have validated the stochastic model by precisely reproducing the established model by Seifert and Beheng (2001) which relies on a simple polynomial kernel. This reproduction matched each equation term for term. The powers of mixing ratio and number concentration are matched in each term, as are each term's coefficient. We've identified a set of stochastic parameters which adhere to conservation of mass and consistency of number and correlate with published values of their model's parameters, thereby facilitating the code match.

In addition to theoretical results, there are empirical results evolving moments of the droplet size spectrum. Using the polynomial kernel that Seifert and Beheng used, we are able to graphically select stochastic parameters that produce a droplet size distribution curve, after 45 minutes of evolution, that precisely matches the evolution curve generated by the well-regarded detailed bin-pair interaction method by Bott (1998). The stochastic model produced reliable results with a time step as large as 30 seconds.

Additionally, for a hydrodynamic and two turbulent kernels, we have been able to graphically select stochastic parameters that convert 50% of cloud mass into rain mass in the same time (to within a few seconds on evolutions that last between 12 minutes and 40 minutes) as droplet size distributions evolved by Bott's LFM (1998). For these additional kernels, we've

collected mean rain radius data that both validates the physical realism of the model and provides droplet evolution characteristics of stochastic parameters.

7.2 Acquiring Parameter Values

7.2.1 A Deterministic Numerical Method

The Reconstruction and Redistribution Algorithm (RRA) uses the mass densities, at the midpoint of the source bins (x_i and x_j) and the midpoints of the two sets of bins (x_{i-1} , x_{i+1} , x_{j-1} and x_{j+1}) which are immediately adjacent to the two source bins, to reconstruct intra-bin quadratic, or linear, distributions of mass density for each source bin. The first three moments of these first or second order polynomial distributions are mass, centroid, and variance. Two sets of three moments readily give the moments of the joint distribution of post collision mass that is distributed over target bins (x_{k-1} , x_k , and x_{k+1}). Moment of the post collision mass are also calculated directly using an assumed distribution of post collision mass density. We thus have two ways of calculating the three moments of the post collision distribution target mass density. Considering the post collision distribution to be quadratic, gives a set of three equations and three unknowns. We integrate the post collision distribution function with integration boundaries determined by the boundaries of the intra-bin distributions in the source bins and by the non-zero domain of the reconstructed distribution in the target bins. Upon normalization, the results of the integration give a percentage of post-collision mass that is added to each target bin. For same bin collisions, this methodology is unnecessary and the entirety of the post-collision mass is added to a single target bin which is determined by the first moment of the joint distribution. The details are given in B.1.

7.2.2 A Stochastic Numerical Method

In 1975, Gillespie presented an algorithm for collision and coalescence of water droplets. His method identified and tracked every particle in the simulation and was therefore quite expensive. In 1996, Seeßelberg et al. modified Gillespie's algorithm by partitioning the droplet size spectrum into bins. This reduced the computational expense because the mass and number in

each bin needed to be tracked instead of tracking every individual particle. However, this modification introduced an error where there were between one and two droplets in a bin. See [Belberg](#) and collaborators did not identify this limitation, nor did they suggest using only integer values of droplets in a unit simulation volume. Using only integer values potentially alters the distribution and may effect the liquid water content of the simulation. To address this limitation without affecting the droplet size distribution, we propose a method to further restrict the sub-interval within a source bin that can be used to select a droplet for collision. The details are given in [B.2](#).

7.2.3 In the Future

We’ve identified four ways to acquire values of stochastic parameters: graphically, using a zero’t order detailed method (Bott’s LFM), using a second order detailed method in [Section 7.2.1](#), and a stochastic detailed method in [Section 7.2.2](#). The last two methods are expensive. The first one lacks the precision we desire, and the second one does not provide the accuracy we want. However, the method we’ve presented in [Section 6.2](#), sequential parameter values, may hold the most promise; and that is the method we propose to employ in future work.

7.3 Broader Impacts

The impact of modelling uncertainty, without (i) any bias that would be inherent in assuming any particular probability distribution, and (ii) the computational expense of performing an ensemble of simulations, will be magnified through the application of this novel technique to additional cloud microphysical processes. More directly, once expanded to include entrainment and detrainment, and validated by observational data, the ‘SDE-based’ stochastic model (i.e. the technique of modelling uncertainty as the sum of a density mean and a set of fluctuations) could be applied to other areas of active research in the climate modelling field that use probability distributions to model unknown quantities such as sea ice, glaciers, and multi-phase cloud microphysics.

Improving the rainfall and radar reflectivity predictions can improve society’s ability to manage risks and mitigate impacts in our changing climate, and

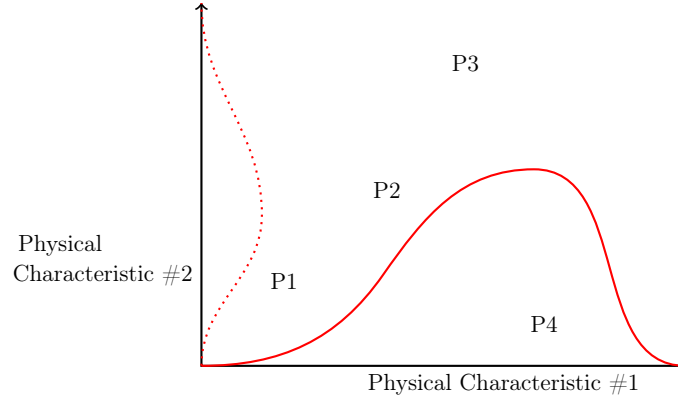
Figure 3.1: Cartoon: Application to Ice Microphysics

Figure 7.1: The axes are a continuum of quantifiable physical characteristics shared by all conventional particles types. Each particle type possesses both characteristics to some degree. P1 is particle #1. P2 is particle #2. P3 is particle #3. P4 is particle #4. The red lines are a probability distributions of the physical characteristics

conveying this information to the public can improve the public’s climate literacy and ability of policy makers to make evidence-based decisions.

7.4 Multi-Phase Cloud Microphysics

Continuing on the path forged by Morrison and Milbrandt (2015), consider a single frozen-water category. This category could contain a variety of conventionally named particles: snow, ice, graupel, and hail. Represent these on a 2D graph with axes that are defined by some quantifiable physical characteristic that is shared by all particles to some degree or another. Examples of such physical characteristics could be (i) mean radial density gradient, (ii) some function of symmetry, (iii) hardness, or (iv) some other distinguishing feature. A cartoon depicting four particles in a 2D continuum is shown in Figure 7.1.

This method would allow frozen water mass to transition smoothly between types of particles rather than as discrete “jumps” from one particle type to another. The probability distributions of these physical characteristics would be represented as (i) density means and (ii) sets of fluctuations. As the distributions of a quantifiable physical characteristic changes, solid water mass would move away from one type of particle and towards another particle

type. This would facilitate the further application of the innovative method developed herein as a model for the evolution of frozen water mass in a developing atmospheric cloud.

8 | Conclusion

Assumptions and simplifications are necessary to produce computationally affordable parameterizations that represent cloud microphysical processes. Parameterizations of cloud microphysical processes over the past forty-five years have made assumptions regarding the droplet size distribution (Seifert & Beheng 2001, Liu & Daum 2004, Liu et al. 2006). Many parameterizations depend on ad-hoc parameters (Khairoutdinov & Kogan 2000, Seifert & Beheng 2001, Liu & Daum 2004, Liu et al. 2006, Franklin 2008). Here, we have derived a stochastic two-moment bulk model of collision and coalescence which does not rely on any specific distribution of the droplet size spectrum, but rather assumes that a distribution exists and that it has a mean. The assumption that any droplet distribution has a mean allows the density to be constructed as the sum of a state space mean and fluctuations from that mean.

The stochastic bulk model is also independent of any specific collision kernel. Our new model can be used with a variety of kernels without any further derivations. The value of the kernel at the mean cloud radius and mean rain radius is used. The flexibility of using any collision kernel comes at the expense of retaining only the zeroth order term in the 2D Taylor expansion of the collision kernel centred at either the cloud, or rain, mean mass. This flexibility requires that the value of the collision kernel be computed, or retrieved from a look-up table, while the stochastic parameterization is being utilized by a climate model.

Existing parameterizations have been restricted to a specific kernel (Seifert & Beheng 2001, Liu & Daum 2004, Liu et al. 2006, Franklin 2008). The kernel in Franklin’s (2008) model was dependent on the turbulent strength, but limited in the range of turbulent kinetic energy used in that kernel.

Seifert and Beheng’s model used a piece-wise polynomial kernel that, with assumptions on the droplet distribution, provided for an analytic solution to the kinetic collection equation. Khairoutdinov and Kogan (2000) calibrated their parameters with data considered to be non-turbulent. Thus their bulk model is limited to representing only gravitational phenomena and their kernel is therefore hydrodynamic.

The substitutions for droplet distribution and collision kernel allow moments of the kinetic collection equation to be computed analytically, save for the higher moments, i.e. the closure problem. The stochastic bulk model uses stochastic processes to close the kinetic collection equation in an unprecedented (to the best of the author’s knowledge) manner. We represent aggregate fluctuations from the state space mean over intervals of the droplet size spectrum as points in a stochastic process. Statistics on these stochastic processes produce the parameters in this new bulk model.

The stochastic bulk model of cloud microphysical processes contains only three stochastic parameters, one for each of three collision processes: cloud-self collection, rain self-collection, and auto conversion. A relationship between the cloud-self collection and auto conversion parameters and the droplet threshold mass allows for an unprecedented controlling of the auto conversion parameter. Auto conversion is the collision and coalescence process most influenced by turbulence, and the one least understood.

The stochastic bulk model was validated against the two-moment bulk model by Seifert and Beheng (2001) that used a piece-wise analytic collision kernel. The validation was theoretical and experimental. The theoretical validation consisted of identifying a region in the 2D parameter space μ_{1n} - μ_{1m} that would precisely reproduce their bulk model, save for the ad-hoc non-dimensional time functions. Experimental validation took the form of choosing values of stochastic parameters that would reproduce evolution curves produced by Bott’s LFM (1998), and choosing other values that would approximate the evolution curves Seifert and Beheng’s bulk model. The stochastic model produced reliable results with a time step as large as 30 seconds.

Stochastic parameters can be chosen such that the evolution by the bulk model reflects particular cloud characteristics, like type (maritime vs. continental and clean vs. polluted) and age. This is particularly important because, with the use of the new stochastic bulk model, a single bulk model of collision and coalescence can be used without the restriction of needing the same droplet size distribution for every cell in a climate model. The new

stochastic bulk model gives the flexibility of choosing different stochastic parameters for different cloud types. Also, different cells in a climate model can use stochastic parameters associated with various collision kernels depending on the intensity and type of turbulence found in the clouds in that climate model cell.

For given values of the three stochastic parameters, the new stochastic parameterization produced results that closely matched results from detailed simulations that used Bott's LFM when the same initial conditions were used. This was done for each of polluted and clean cloud types. A simple metric of the time required to convert half of the cloud mixing ratio to rain mixing was used to validate the results of the stochastic bulk model. It is demonstrated here that while adequate parameter values be found for the stochastic model, they can depend heavily of the initial cloud mean radius, i.e, the environmental conditions or the cloud's age. Moreover, for the two experiments considered here, despite the utilization of an ad hoc adjustment function to correct excessive accretion and auto-conversion rates, Siefert and Beheng's model either overestimate or underestimate the time at which 50% of the cloud mass is converted into rain and the stochastic parameters that are found to reproduce faithfully the detailed KCE results can sometimes lies outside the region of validity constrained by Siefert and Beheng parameters. This, in essence, demonstrates the usefulness of the stochastic bulk parameterization.

The sensitivity of the "50% conversion time" was examined as a function of the 2D μ_{1n} - μ_{1m} space. This conversion time metric was sensitive to small changes in the auto conversion μ_{1m} parameter near the values that would reproduce the evolution curve produced by the detailed method, but showed very little sensitivity elsewhere in the domain of physically permissible values. The conversion time metric showed little sensitivity to the cloud self-collection μ_{1n} parameter anywhere on this domain for a given value of the rain self-collection μ_{4np} parameter. The mean rain radius at the time of 50% conversion was sensitive to the value of the rain self-collection parameter. Both the conversion time and the mean rain radius at 50% conversion time showed very little sensitivity to changes in the time step in the range 0.1-30 seconds.

The stochastic bulk model was extended to show its utility with hydrodynamic and turbulent collision kernels. A parameterized turbulent kernel by Franklin (2008) enhanced collision rates for droplets with a radii between 10-30 μm over hydrodynamic collision rates and most prominently enhanced

clustering phenomena. A tabulated turbulent kernel developed by Pinsky and Khain (2008) enhanced collision rates for droplets with a radii between 1-21 μm . Their kernel most sharply enhanced phenomena governed by fluid velocity differentials. The stochastic bulk model used the strongest turbulence allowed by each of these turbulent formulations. The hydrodynamic kernel was used for droplets outside of these ranges.

For each of these three kernels, simulations were performed that represented a clean and a polluted cloud. For each of the six simulations, results were collected that gave the time required to convert 50% of cloud mass to rain mass, the mean rain radius at that time, and the initial mean rain radius. These results were collected over a region in the 2D μ_{1n} - μ_{1m} space that was subject to a controlled auto conversion parameter. The 50% conversion time was compared to results from Bott's LFM, and in the case of the hydrodynamic kernel to Khairoutdinov and Kogan's model. In each case stochastic parameters could be located that reproduced the 50% conversion time of the detailed method for the respective initial conditions and choice of kernel.

The collection of simulations using the hydrodynamic and turbulent kernels showed a number of consistencies between the kernels. The initial mean rain radius over the region of validity in the μ_{1n} - μ_{1m} space has a smooth transition from the smallest possible mean rain radius (40 μm) to the largest possible (50 μm) for all simulations except for the clean cloud driven by a tabulated kernel. In that case, the initial mean rain radius was constant except for a small corner of the contour plot. This could be due to the closeness of the initial mean cloud radius to the threshold radius. The mean rain radius at the time of 50% mass conversion gave expected results. The stochastic parameters that produced a larger initial mean rain radius also produced a larger mean rain radius at the time of 50% mass conversion.

Of interest to note is that the contour lines of constant 50% conversion time changed orientation as the orientation of the triangular region containing valid parameter values changed. The orientation of the triangle is governed by the relationship of the threshold mass to the mean cloud mass. When $x^*/\bar{x}_c$ is large, ≈ 64 , the contour lines are horizontal i.e. the 50% conversion time is sensitive to the auto conversion parameter but not the cloud self-collection parameter. As $x^*/\bar{x}_c$ decreases, the contour lines shift such that the 50% conversion time is more sensitive to the cloud self-collection parameter and less sensitive to the auto conversion parameter. When $x^*/\bar{x}_c \approx 4$, we see very little dependence on the auto conversion parameter and maximal

dependence on the cloud self-collection parameter.

Stochastic parameter values were obtained using Bott’s (1998) detailed method for all six simulations. We saw that none of the simulations produced values for the parameters that would reasonably approximate results from the detailed method for the entire evolution of the droplet spectrum. This may be due to the approximations inherent in the discretization scheme that uses the midpoint of a bin to discretize a continuous distribution function. We began initial work into two other methods of acquiring values for the stochastic parameters: a quadratically-based detailed method and a stochastic simulation (Gillespie 1975*a*, Seesselberg et al. 1996). The quadratic method was an order of magnitude more expensive computationally. The stochastic simulation method is also expensive, but may eliminate the bias associated with two nested FOR-loops in a detailed scheme as are in Bott’s LFM (1998).

This work can impact the way cloud microphysical parameterizations are used by climate models. Once activation, condensation, evaporation, and entrainment/detrainment are included, a comprehensive stochastic model can allow climate models to implement various collision kernels without replacing the microphysics scheme and without limiting to a particular droplet size distribution. Our method of using stochastic processes to close the kinetic collection equation may be exported to other areas that need moment closure. Finally, our method of using the physics of the phenomena to reshape state space fluctuations into fluctuations in a stochastic process may have applications to situations where uncertainty needs to be quantified for effective modelling, but the probability distribution associated with that uncertainty is currently under debate, perhaps with sea ice, glaciers, or extreme events.

Future work can include using these methods to model the uncertainty associated with entrainment/detrainment. In 2013 Dawe and Austin used LES to determine probability distribution functions for entrainment and detrainment rates. They showed various cloud core properties having normal, exponential, and log-normal distributions with a dependence on buoyancy characteristics (Dawe & Austin 2013). In other entrainment models, Poisson distributions (Suseli, Teixeira & Chung 2013) and uniform (Romps & Kuang 2010, Raymond & Blyth 1986) have been used for entrainment rates, exponential for plume size (Suseli et al. 2013), and Gaussian for thermal updraft properties (Romps & Kuang 2010). Nevertheless, Suseli and collaborators (2013) state that entrainment rates don’t allow enough variability. However, Romps and Kuang (2010) showed that variability at cloud base

is inconsequential relative to lateral entrainment. The distribution of moist conserved variables and vertical velocity deviate from a normal distribution (Suseli et al. 2013). In a 2013 review paper, de Rooy and collaborators identified spatial and temporal variations in cloud properties to be a reason to avoid using averaged quantities and ad-hoc parameters and stated that models of these mixing processes are still in infancy (de Rooy et. al. 2013)

Future work can also include finding a suitable method of acquiring values for the stochastic parameters. Further developing a method associated with stochastic simulations seems more likely than developing a higher order detailed method given the sequencing bias associated with the latter. It may be possible to use variations on a simple and computationally inexpensive detailed method, such as acquiring a parameter value when cloud self-collection is dominant and another value when accretion is dominant. Other possible methods would be to use a Markov jump process to sample the region of valid stochastic parameters that would be conditional on environmental conditions (e.g. clean v.s. polluted, marine v.s. continental) and the age of the cloud (Khouider et. al., 2010) , or to parameterize selection in the 2D μ_{1n} - μ_{1m} space conditional on entrainment/detrainment and nucleation processes.

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A | Appendix

A.1 Hydrodynamic Kernel Values

Table A.1: Hydrodynamic Collision Efficiencies (6-40 μm)

Radius Ratio	[non-dimensional]							
	Collector Drop Radius [μm]							
	6	8	10	15	20	25	30	40
0.00	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.05	0.003	0.003	0.004	0.005	0.005	0.005	0.008	0.020
0.10	0.005	0.005	0.006	0.007	0.008	0.009	0.010	0.070
0.15	0.010	0.010	0.010	0.011	0.012	0.016	0.030	0.280
0.20	0.012	0.012	0.012	0.015	0.020	0.028	0.050	0.500
0.25	0.017	0.017	0.017	0.020	0.022	0.053	0.100	0.620
0.30	0.020	0.020	0.020	0.022	0.032	0.062	0.200	0.680
0.35	0.022	0.022	0.022	0.027	0.043	0.107	0.265	0.740
0.40	0.027	0.027	0.027	0.031	0.052	0.152	0.330	0.780
0.45	0.030	0.030	0.030	0.034	0.064	0.196	0.395	0.800
0.50	0.033	0.033	0.033	0.037	0.068	0.240	0.460	0.800
0.55	0.035	0.035	0.035	0.038	0.079	0.240	0.460	0.800
0.60	0.037	0.037	0.037	0.039	0.082	0.245	0.465	0.780
0.65	0.038	0.038	0.038	0.040	0.080	0.250	0.470	0.770
0.70	0.037	0.037	0.037	0.041	0.075	0.250	0.470	0.760
0.75	0.037	0.037	0.037	0.039	0.067	0.202	0.385	0.770
0.80	0.036	0.036	0.036	0.037	0.057	0.154	0.300	0.770
0.85	0.035	0.035	0.035	0.037	0.048	0.106	0.215	0.780
0.90	0.035	0.035	0.035	0.037	0.040	0.058	0.130	0.790
0.95	0.029	0.029	0.029	0.033	0.033	0.058	0.130	0.950
1.00	0.027	0.027	0.027	0.027	0.027	0.058	0.130	1.400

Table A.1: Collision Efficiencies by Hall (1980) modified to reproduce the contour plot from Seibelberg (1996).

Table A.2: Hydrodynamic Collision Efficiencies (50-300 μm)
[non-dimensional]

Radius Ratio	50	60	70	100	150	200	300
0.00	0.000	0.001	0.001	0.001	0.001	0.001	0.001
0.05	0.080	0.150	0.200	0.500	0.770	0.870	0.970
0.10	0.400	0.430	0.580	0.790	0.930	0.960	1.000
0.15	0.600	0.640	0.750	0.910	0.970	0.980	1.000
0.20	0.700	0.770	0.840	0.950	0.970	1.000	1.000
0.25	0.780	0.840	0.880	0.950	1.000	1.000	1.000
0.30	0.830	0.870	0.900	1.000	1.000	1.000	1.000
0.35	0.860	0.890	0.920	1.000	1.000	1.000	1.000
0.40	0.880	0.900	0.940	1.000	1.000	1.000	1.000
0.45	0.900	0.910	0.950	1.000	1.000	1.000	1.000
0.50	0.900	0.910	0.950	1.000	1.000	1.000	1.000
0.55	0.900	0.910	0.950	1.000	1.000	1.000	1.000
0.60	0.900	0.910	0.950	1.000	1.000	1.000	1.000
0.65	0.890	0.910	0.950	1.000	1.000	1.000	1.000
0.70	0.880	0.920	0.950	1.000	1.000	1.000	1.000
0.75	0.880	0.930	0.970	1.000	1.000	1.000	1.000
0.80	0.890	0.950	1.000	1.000	1.000	1.000	1.000
0.85	0.920	1.000	1.020	1.020	1.020	1.020	1.020
0.90	1.010	1.030	1.040	1.040	1.040	1.040	1.040
0.95	1.300	1.700	2.300	2.300	2.300	2.300	2.300
1.00	2.300	3.000	4.000	4.000	4.000	4.000	4.000

Table A.2: Collision Efficiencies by Hall (1980) modified to reproduce the contour plot from See β elberg (1996).

A.2 Tabulated Turbulent Kernel Values

Table A.3: Turbulent Kernel Enhancement Factors (1-7 μm)
[non-dimensional]

Radius	Source Drop Radius [μm]						
	1	2	3	4	5	6	7
1	9.5088	10.7165	7.7190	7.3119	7.4381	7.7744	8.0197
2	10.7165	9.5088	8.3012	5.2668	4.4904	4.1161	3.9747
3	7.7190	8.3012	8.0510	7.8009	4.7712	3.8168	3.4314
4	7.3119	5.2668	7.8009	7.6910	7.5812	4.5783	3.6268
5	7.4381	4.4904	4.7712	7.5812	7.5398	7.4984	4.52580
6	7.7744	4.1161	3.8168	4.5783	7.4984	7.4897	7.4810
7	8.0197	3.9747	3.4314	3.6268	4.5258	7.4810	7.5203
8	8.4033	3.9324	3.2362	3.2758	3.5906	4.5186	7.5597
9	8.9368	3.9381	3.1702	3.1392	3.3689	3.7252	4.6303
10	9.8044	4.0199	3.2251	3.2509	3.4602	3.7232	4.0957
11	10.3258	4.1629	3.3740	3.5154	3.8244	4.0518	4.2680
12	11.1223	4.3113	3.5160	3.8466	4.3378	4.6825	4.8944
13	11.6593	4.4118	3.8447	4.3818	5.0158	5.4507	5.7149
14	12.5091	4.6412	4.1504	5.0138	5.8742	6.3310	6.5808
15	12.9058	4.8549	4.6054	5.7319	6.7182	7.2524	7.4753
16	14.2514	5.1548	5.0845	6.6208	7.6810	8.1286	8.1837
17	15.0638	5.3733	5.7038	7.5365	8.7647	8.9361	8.6637
18	16.0748	5.8802	6.5236	8.8114	9.8164	9.6136	8.7714
19	16.1598	6.3539	7.3636	9.8931	10.9418	10.1262	8.5535
20	17.5126	6.6672	8.3580	11.3477	11.9506	10.3358	7.9211
21	17.9564	7.3990	9.5677	12.7411	13.0221	10.3699	7.2376

Table A.3: Turbulent Kernel Enhancement Factors by Pinsky and Khain (2008).
Turbulent Kinetic Energy is $1000 \text{ cm}^2\text{s}^{-3}$ and the Reynolds number is 20,000.

Table A.4: Turbulent Kernel Enhancement Factors (8-14 μm)
[non-dimensional]

Source Drop Radius [μm]							
Radius	8	9	10	11	12	13	14
1	8.4033	8.9368	9.8044	10.3258	11.1223	11.6593	12.5091
2	3.9324	3.9381	4.0199	4.1629	4.3113	4.4118	4.6412
3	3.2362	3.1702	3.2251	3.3740	3.5160	3.8447	4.1504
4	3.2758	3.1392	3.2509	3.5154	3.8466	4.3818	5.0138
5	3.5906	3.3689	3.4602	3.8244	4.3378	5.0158	5.8742
6	4.5186	3.7252	3.7232	4.0518	4.6825	5.4507	6.3310
7	7.5597	4.6303	4.0957	4.2680	4.8944	5.7149	6.5808
8	7.5445	7.5294	5.0570	4.5791	4.9908	5.8047	6.7272
9	7.5294	8.1570	8.7845	5.5385	5.2224	5.8259	6.7751
10	5.0570	8.7845	8.6383	8.4920	5.7163	5.7524	6.5562
11	4.5791	5.5385	8.4920	8.6152	8.7384	6.1729	6.3671
12	4.9908	5.2224	5.7163	8.7384	8.8907	9.0431	6.6560
13	5.8047	5.8259	5.7524	6.1729	9.0431	9.2233	9.4035
14	6.7272	6.7751	6.5562	6.3671	6.6560	9.4035	9.5439
15	7.5814	7.6070	7.4326	7.2449	7.0821	7.2749	9.6843
16	8.0784	8.0032	7.8985	7.8958	7.8082	7.6762	7.8707
17	8.2338	7.8228	7.6538	7.7381	7.9285	8.1984	8.2404
18	7.8285	7.0866	6.7328	6.6962	6.9644	7.5963	8.2333
19	7.0592	6.1694	5.6604	5.5343	5.6541	6.0840	6.8451
20	6.2194	5.3303	4.8257	4.6300	4.6461	4.8376	5.2457
21	5.5028	4.6692	4.2467	4.0466	3.9769	4.0461	4.2380

Table A.4: Turbulent Kernel Enhancement Factors by Pinsky and Khain (2008).
Turbulent Kinetic Energy is $1000 \text{ cm}^2\text{s}^{-3}$ and the Reynolds number is 20,000.

Table A.5: Turbulent Kernel Enhancement Factors (15-21 μm)
[non-dimensional]

Radius	Source Drop Radius [μm]						
	15	16	17	18	19	20	21
1	12.9058	14.2514	15.0638	16.0748	16.1598	17.5126	17.9564
2	4.8549	5.1548	5.3733	5.8802	6.3539	6.6672	7.3990
3	4.6054	5.0845	5.7038	6.5236	7.3636	8.3580	9.5677
4	5.7319	6.6208	7.5365	8.8114	9.8931	11.3477	12.7411
5	6.7182	7.6810	8.7647	9.8164	10.9418	11.9506	13.0221
6	7.2524	8.1286	8.9361	9.6136	10.1262	10.3358	10.3699
7	7.4753	8.1837	8.6637	8.7714	8.5535	7.9211	7.2376
8	7.5814	8.0784	8.2338	7.8285	7.0592	6.2194	5.5028
9	7.6070	8.0032	7.8228	7.0866	6.1694	5.3303	4.6692
10	7.4326	7.8985	7.6538	6.7328	5.6604	4.8257	4.2467
11	7.2449	7.8958	7.7381	6.6962	5.5343	4.6300	4.0466
12	7.0821	7.8082	7.9285	6.9644	5.6541	4.6461	3.9769
13	7.2749	7.6762	8.1984	7.5963	6.0840	4.8376	4.0461
14	9.6843	7.8707	8.2404	8.2333	6.8451	5.2457	4.2380
15	9.9194	10.1545	8.5332	8.7228	7.9797	6.0779	4.6300
16	10.1545	10.4820	10.8096	9.1521	9.1338	7.4376	5.3751
17	8.5332	10.8096	11.1228	11.4361	9.9598	9.2427	6.7770
18	8.7228	9.1521	11.4361	11.8981	12.3602	10.6858	9.0917
19	7.9797	9.1338	9.9598	12.3602	12.9910	13.6218	11.3308
20	6.0779	7.4376	9.2427	10.6858	13.6218	14.2508	14.8798
21	4.6300	5.3751	6.7770	9.0917	11.3308	14.8798	14.2508

Table A.5: Turbulent Kernel Enhancement Factors by Pinsky and Khain (2008).
Turbulent Kinetic Energy is $1000 \text{ cm}^2\text{s}^{-3}$ and the Reynolds number is 20,000.

A.3 Moments of the Source Bin Distributions

Table A.6: Reconstruction Polynomial: First Moment, Primary

Eqn.	Polynomial Form	First Moment, $\overline{x_i^*} =$
(B.6a)	Quadratic	$\frac{a}{g_i} \left[(2 - 2y_{i+di/2} + y_{i+di/2}^2) \exp(y_{i+di/2}) - \right.$
(B.6b)		$\left. (2 - 2y_{i-di/2} + y_{i-di/2}^2) \exp(y_{i-di/2}) \right] +$ $\frac{b}{g_i} \left[(y_{i+di/2} - 1) \exp(y_{i+di/2}) - \right.$ $\left. (y_{i-di/2} - 1) \exp(y_{i-di/2}) \right] +$ $\frac{c}{g_i} \left[\exp(y_{i+di/2}) - \exp(y_{i-di/2}) \right]$
(B.8)	First bin linear $i = 1$	$\frac{p_1}{g_1} \left[y_{1+1/2} \exp(y_{1+1/2}) - y_{1/2} \exp(y_{1/2}) - \right.$ $\left. \exp(y_{1+1/2}) + \exp(y_{1/2}) \right] +$ $\frac{q_1}{g_1} \left[\exp(y_{1+1/2}) - \exp(y_{1/2}) \right]$
(B.11)	Last bin linear $i = n$	$\frac{p_n}{g_n} \left[y_{n+1/2} \exp(y_{n+1/2}) - y_{n-1/2} \exp(y_{n-1/2}) - \right.$ $\left. \exp(y_{n+1/2}) + \exp(y_{n-1/2}) \right] +$ $\frac{q_n}{g_n} \left[\exp(y_{n+1/2}) - \exp(y_{n-1/2}) \right]$

Table A.6: The first moment of the reconstruction polynomial

Table A.7: Reconstruction Polynomial: First Moment, Alternate

Eqn.	Polynomial Form	First Moment, $\overline{x_i^*} =$
(B.18)	3-piece alternative linear (mass on both sides of bin)	$\frac{\alpha}{g_i} \left[\exp(y_{i-1/2})(y_{i-1/2} - 1 - y_{r1}) + \exp(y_{r1}) \right] +$ $\frac{\beta}{g_i} \left[\exp(y_{i+1/2})(y_{i+1/2} - 1 - y_{r2}) + \exp(y_{r2}) \right]$
(B.20)	2-piece alternative. linear (mass on left side of bin)	$\frac{\alpha}{g_i} \left[\exp(y_{i-1/2})(y_{i-1/2} - 1 - y_{r1}) + \exp(y_{r1}) \right]$
(B.21)	2-piece alternative. linear (mass on right side of bin)	$\frac{\beta}{g_i} \left[\exp(y_{i+1/2})(y_{i+1/2} - 1 - y_{r2}) + \exp(y_{r2}) \right]$

Table A.7: The first moment of the reconstruction polynomial

Table A.8: Reconstruction Polynomial: Second Moment, Primary

Eqn.	Polynomial Form	Second Moment, $\overline{x_i^2} =$
(B.6a)	Quadratic	$\frac{a}{4g_i} \left[(1 - 2y_{i+di/2} + 2y_{i+di/2}^2) \exp(2y_{i+di/2}) - \right.$
(B.6b)		$\left. (1 - 2y_{i-di/2} + 2y_{i-di/2}^2) \exp(2y_{i-di/2}) \right] +$ $\frac{b}{2g_i} \left[(y_{i+di/2} - 0.5) \exp(2y_{i+di/2}) - \right.$ $\left. ((y_{i-di/2} - 0.5) \exp(2y_{i-di/2})) \right] +$ $\frac{c}{2g_i} \left[\exp(2y_{i+di/2}) - \exp(2y_{i-di/2}) \right]$
(B.8)	First bin linear $i = 1$	$\frac{p_1}{2g_1} \left[y_{1+1/2} \exp(2y_{1+1/2}) - y_{1/2} \exp(2y_{1/2}) - \right.$ $\left. 0.5 \exp(2y_{1+1/2}) + 0.5 \exp(2y_{1/2}) \right] +$ $\frac{q_1}{2g_1} \left[\exp(2y_{1+1/2}) - \exp(2y_{1/2}) \right]$
(B.11)	Last bin linear $i = n$	$\frac{p_n}{2g_n} \left[y_{n+1/2} \exp(2y_{n+1/2}) - y_{n-1/2} \exp(2y_{n-1/2}) - \right.$ $\left. 0.5 \exp(2y_{n+1/2}) + 0.5 \exp(2y_{n-1/2}) \right] +$ $\frac{q_n}{2g_n} \left[\exp(2y_{n+1/2}) - \exp(2y_{n-1/2}) \right]$

Table A.8: The second moment of the reconstruction polynomial

Table A.9: Reconstruction Polynomial: Second Moment, Alternate

Eqn.	Polynomial Form	Second Moment, $\overline{x_i^2} =$
(B.18)	3-piece alternative linear (mass on both sides of bin)	$\frac{\alpha}{2g_i} \left[\exp(2y_{i-1/2})(y_{i-1/2} - 0.5 - y_{r1}) + 0.5 \exp(2y_{r1}) \right]$ $\frac{\beta}{2g_i} \left[\exp(2y_{i+1/2})(y_{i+1/2} - 0.5 - y_{r2}) + 0.5 \exp(2y_{r2}) \right]$
(B.20)	2-piece alternative linear (mass on left side of bin)	$\frac{\alpha}{2g_i} \left[\exp(2y_{i-1/2})(y_{i-1/2} - 0.5 - y_{r1}) + 0.5 \exp(2y_{r1}) \right]$
(B.21)	2-piece alternative linear (mass on right side of bin)	$\frac{\beta}{2g_i} \left[\exp(2y_{i+1/2})(y_{i+1/2} - 0.5 - y_{r2}) + 0.5 \exp(2y_{r2}) \right]$

Table A.9: The second moment of the reconstruction polynomial

A.4 Compatibility Condition

For any given droplet size of mass x , the number concentration density and the mixing ratio density at time t are $n(x, t)$ and $q(x, t)$, respectively; and are related by

$$q(x, t) = xn(x, t).$$

Using the second relation in Equation (3.1) and the physical relation between the two densities, another expression for aggregate cloud mixing ratio is

$$q_c(t) = \int_0^{x^*} q(x, t) dx.$$

The two compatibility conditions are

$$\int_0^{x^*} xn(x, t) dx = \int_0^{x^*} q(x, t) dx \quad \text{and} \quad \lim_{x_m \rightarrow \infty} \int_{x^*}^{x_m} xn(x, t) dx = \lim_{x_m \rightarrow \infty} \int_{x^*}^{x_m} q(x, t) dx$$

Cloud Aggregate Compatibility

The cloud aggregate approximation gives

$$\int_0^{x^*} x \left(\frac{N_c(t)}{x^*} + \delta_c(\omega_1; x, t) \right) dx = \int_0^{x^*} \frac{q_c(t)}{x^*} + \gamma_c(\omega_2; x, t) dx.$$

Integrating the deterministic parts produces

$$\frac{x^* N_c(t)}{2} + \int_0^{x^*} x \delta_c(\omega_1; x, t) dx = q_c(t) + \int_0^{x^*} \gamma_c(\omega_2; x, t) dx.$$

The integral on the right hand side is shown to be zero in Section (3.2), and the end of the proof is given in Equation (3.8). A slight rearrangement gives

$$\int_0^{x^*} x \delta_c(\omega_1; x, t) dx = q_c(t) - \frac{x^* N_c(t)}{2} = N_c \left(\bar{x}_c - \frac{x^*}{2} \right) \quad (\text{A.1})$$

The relationship between the centre of mass of the cloud aggregate and the separation mass shows that the weighted number fluctuation within the cloud aggregate is negative when $2\bar{x}_c \leq x^*$. The negative values for the

integral exist for most of the cloud evolution, and this can be verified with data.

Rain Aggregate Compatibility

The rain aggregate approximation gives

$$\lim_{x_m \rightarrow \infty} \int_{x^*}^{x_m} x \left(\frac{N_r(t)}{x_m - x^*} + \delta_r(\omega_1; x, t) \right) dx = \lim_{x_m \rightarrow \infty} \int_{x^*}^{x_m} \frac{q_r(t)}{x_m - x^*} + \gamma_c(\omega_2; x, t) dx.$$

which produces the following expression for the weighted number fluctuation within the rain aggregate

$$\lim_{x_m \rightarrow \infty} \int_{x^*}^{x_m} x \delta_r(\omega_1; x, t) dx = q_r(t) - \lim_{x_m \rightarrow \infty} \frac{(x_m + x^*) N_r(t)}{2}.$$

Because droplets cannot grow to an infinite mass (Pruppacher & Klett 1997), take $x_m \gg x^*$ to be finite and have an expression for the weighted number fluctuation within the rain aggregate that can be acquired numerically:

$$\int_{x^*}^{x_m} x \delta_r(\omega_1; x, t) dx = q_r(t) - \frac{x_m N_r(t)}{2} = N_r \left(\bar{x}_r - \frac{x_m}{2} \right). \quad (\text{A.2})$$

The relationship between the centre of mass of the rain aggregate and the separation mass shows that the weighted number fluctuation within the rain aggregate is negative when $2\bar{x}_r \leq x^*$. The negative values for the integral exist for the entirety of the cloud evolution, and this can be also verified with data.

In the current bulk cloud microphysics parameterization developed herein, the mass term which is added to the kinetic collection equation given by Equation 2.1 to get the first moment is absorbed by $q(x, t) = xn(x, t)$. Therefore the left hand side of Equations (A.1 and A.2) do not appear in the derivations nor the final stochastic parameterization, and these compatibility conditions do not constrain the parameterization developed herein.

Any stochastic parameterization which uses the number concentration density approximation in Equation 3.3 and omits the mixing ratio density approximation in Equation 3.4 will contain the left hand side of Equations (A.1 and A.2) in the derivations and the final parameterization. Thus the utility of those parameterizations will be limited by conditions imposed by Equations (A.1 and A.2).

A.5 Stochastic Parameter Nomenclature

The suite of indices used in the forty-two stochastic parameters can be partitioned into three classes. The largest class uses three alpha numeric characters. The other two classes have either two or four characters. In all three classes the first character takes values between one and six and indicates a region in the pre-collision droplet domain shown in Figure (3.1).

The class with four characters is used to index stochastic processes modelling the accretion process. The second and third characters are either *cn*, *cm*, *rn*, or *rm* and represent cloud number, rain number, cloud mixing ratio, and rain mixing ratio, respectively. This distinction is necessary only for the accretion process because none of the three accretion regions in Figure (3.1) are symmetric with respect to the $y = z$ line.

The class with three characters describe stochastic processes in the remaining collision and coalescence processes, and these occur in the regions in Figure (3.1) that are symmetric about the line $y = z$. The class with two characters identify stochastic processes that are linear combinations of processes with three or four characters. The indices for two stochastic processes are detailed:

$$\mu_{4nc} = \lim_{x_m \rightarrow \infty} \frac{1}{x^* N_r^2} \int_{\Omega_4} \delta_r(y) \delta_r(z) dz dy$$

$$\mu_{3cmf} = \lim_{x_m \rightarrow \infty} \frac{1}{(x_m - x^*) q_c} \int_{\Omega_3} \gamma_c(y) dz dy$$

The index in the first process identifies ‘the covariance of number concentration fluctuations in region 4.’ The index in the second process identifies ‘the fluctuations of cloud mixing ratio in region 3.’

B | Supplementary Material

B.1 A Detailed Numerical Method for the KCE

B.1.1 Source Bin Distributions

In this section we describe recent work towards a 2nd Order Detailed Method for numerically discretizing the KCE. In each simulated collision, two probability density functions (pdf), $m(y_i)$ and $m(y_j)$, are constructed, where $y_i = \ln(x_i)$. Each pdf represents the distribution of mass density within bins x_i and x_j , respectively. Distinct quadratic interpolations is used for each bin except for the first and last bins. A linear interpolation is used to reconstruct the pdf of mass density within the first and last bins. The quadratic pdf's are constructed using the total mass density of a bin, say bin x_i , and its two immediate neighbouring bins: x_{i-1} and x_{i+1} . The linear probability distribution functions for the 1st and last bins are constructed using the total mass densities of bins x_1 and x_2 or, for the case of n bins, the total mass densities of bins x_{n-1} and x_n . For the quadratic interpolation, the total mass densities of these three bins are g_i, g_{i-1} , and g_{i+1} , respectively, and each is clearly positive. For the linear interpolation, the total mass densities are g_1 and g_2 or g_n and g_{n-1} for the first and last bins, respectively. The pdf of the distribution of droplets within bin x_j , is calculated using a similar procedure.

If $m(y_i)$ becomes negative anywhere in bin x_i , then the reconstruction does not represent a physical distribution of mass density (Tzivion et al. 1987). There are particular combinations of g_i, g_{i-1} , and g_{i+1} that will cause the reconstruction polynomial to have negative values in bin x_i . Similarly, there

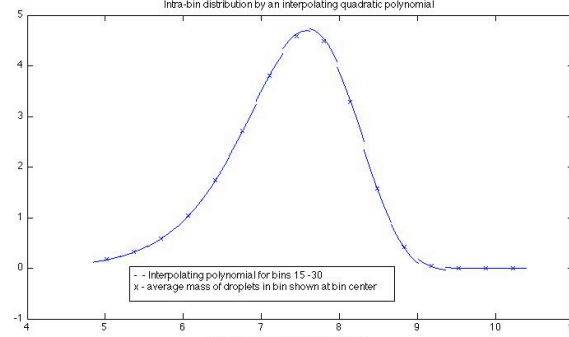
Figure B.1: Source Bins: Multiple Intra-Bin Distribution

Figure B.1: A example of a collection of quadratic distributions. The abscissa is the non-dimensional log-radius and the ordinate is the mass density.

are combinations of g_1 and g_2 , or g_{n-1} and g_n that will cause $m(y_i)$ to become negative somewhere in bin x_1 or bin x_n . When either reconstruction polynomial takes negative values in the respective bin, the pdf of the mass density represented by that polynomial needs to be reconstructed using an alternative form.

We use the roots of the reconstruction parabola, or the root of the linear interpolating polynomial to construct a piece-wise alternate linear pdf of mass density. The mass densities that had negative values given by the polynomial are set to zero. The remaining mass density is adjusted to conserve mass. This alternate reconstruction gives a piece-wise linear polynomial as the mass density function within the bin. The polynomial reconstruction algorithm uses either a parabolic, linear, or an alternate piece-wise linear function to distribute the mass density within the bin.

We derive the quadratic, linear, and alternate piece-wise reconstructions, and emphasize that g_i is derived from the *inter*-bin distribution of mass density and used to construct an *intra*-bin distribution of mass density. In the cases where $i = j \pm 1$ the distributions for bin i and bin j share two points of interpolation and when $i = j \pm 2$ they share a single point. However, the distributions are considered to be dependent only when they share all three points of interpolation.

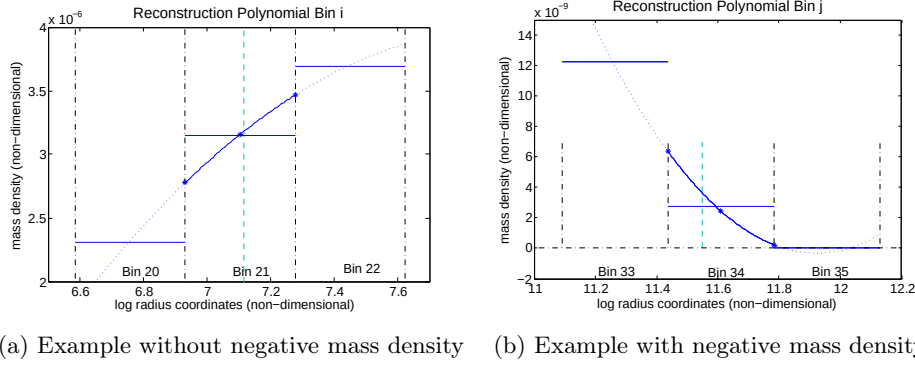
Figure B.2: Source Bins: Quadratic Distribution

Figure B.2: Both examples have an initial distribution with a liquid water content of 1g cm^{-3} and a number concentration of 239 cm^{-3} . The horizontal blue lines are constant intra-bin distributions of mass density. The curved blue lines are quadratic approximation of mass density for Bins 21 and 34. The black dotted-dashed lines are bin boundaries, and the aqua lines (and blue dots) are centroids (and mid & end points) of the quadratic distributions, respectively.

Quadratic Reconstruction

The total mass densities, g_i, g_{i-1} , and g_{i+1} are known from the distribution of mass density *between* the bins and are first given by the initial gamma distribution and subsequently given by an evolved form of that distribution. Assuming the same distribution, $m(y)$, within all three adjacent bins, we consider a polynomial distribution of the mass density and use the following three density formulas.

$$\int_{y_{i-1}-\frac{\delta(i-1)}{2}}^{y_{i-1}+\frac{\delta(i-1)}{2}} m(\hat{y})d\hat{y} = g_{i-1}, \quad \int_{y_i-\frac{\delta i}{2}}^{y_i+\frac{\delta i}{2}} m(\hat{y})d\hat{y} = g_i, \quad \int_{y_{i+1}-\frac{\delta(i+1)}{2}}^{y_{i+1}+\frac{\delta(i+1)}{2}} m(\hat{y})d\hat{y} = g_{i+1}, \quad (\text{B.1})$$

where δi is the width of bin i , and $m(y_i)$ is a quadratic form of the mass density function within bin x_i :

$$m(y_i) = ay_i^2 + by_i + c. \quad (\text{B.2})$$

Since the bins are adjacent, $y_{i-\frac{\delta i}{2}} = y_{i-1}+\frac{\delta(i-1)}{2}$ and $y_{i+\frac{\delta i}{2}} = y_{i+1}-\frac{\delta(i+1)}{2}$. Two examples of a quadratic interpolation are presented in Figure B.2. Inte-

grating the same quadratic polynomial given as Equation B.2 in each of the three formulas in Equation B.1, gives a system of three equations in three unknowns.

$$\begin{aligned} \frac{1}{3} \left[y_{i-\frac{\delta i}{2}}^3 - y_{\gamma(i-1)}^3 \right] a + \frac{1}{2} \left[y_{i-\frac{\delta i}{2}}^2 - y_{\gamma(i-1)}^2 \right] b + \left[y_{i-\frac{\delta i}{2}} - y_{\gamma(i-1)} \right] c &= g_{i-1} \\ \frac{1}{3} \left[y_{i+\frac{\delta i}{2}}^3 - y_{i-\frac{\delta i}{2}}^3 \right] a + \frac{1}{2} \left[y_{i+\frac{\delta i}{2}}^2 - y_{i-\frac{\delta i}{2}}^2 \right] b + \left[y_{i+\frac{\delta i}{2}} - y_{i-\frac{\delta i}{2}} \right] c &= g_i \\ \frac{1}{3} \left[y_{\gamma(i+1)}^3 - y_{i+\frac{\delta i}{2}}^3 \right] a + \frac{1}{2} \left[y_{\gamma(i+1)}^2 - y_{i+\frac{\delta i}{2}}^2 \right] b + \left[y_{\gamma(i+1)} - y_{i+\frac{\delta i}{2}} \right] c &= g_{i+1} \end{aligned}$$

where $\gamma(i-1) = i-1 - \frac{\delta(i-1)}{2}$ and $\gamma(i+1) = i+1 + \frac{\delta(i+1)}{2}$.

Substituting Y_{1m} for $\frac{1}{3} \left(y_{i-\frac{\delta i}{2}}^3 - y_{\gamma(i-1)}^3 \right)$ and making the eight other corresponding substitutions simplifies the system of equations.

In Equation B.4, the indices ‘ m ’ and ‘ p ’ are abbreviations for minus and plus and refer to the indexing of the bins, i.e. the index ‘ m ’ refers to the lower bin boundary such as in y_{i-1} . The indices 1, 2, and 3 simply refers to the ordinality of the terms in the left-hand-side of Equation B.4. This choice of indexing is intended to reduce confusion with a similar linear system presented in Section B.1.3.

$$\begin{aligned} Y_{1m}a + Y_{2m}b + Y_{3m}c &= g_{i-1} \\ Y_{1a} + Y_{2b} + Y_{3c} &= g_i \\ Y_{1p}a + Y_{2p}b + Y_{3p}c &= g_{i+1} \end{aligned} \tag{B.4}$$

Solving the system of equations given in Equation B.4 for a , b , and c gives the coefficients of the quadratic form of the mass density function, Equation B.2 for source bin i :

$$\begin{aligned} a &= \frac{(Y_{2p}Y_3 - Y_2Y_{3p})g_{i-1} + (Y_{2m}Y_{3p} - Y_{2p}Y_{3m})g_i + (Y_2Y_{3m} - Y_{2m}Y_3)g_{i+1}}{\Omega} \\ b &= \frac{(Y_1Y_{3p} - Y_{1p}Y_3)g_{i-1} + (Y_{1p}Y_{3m} - Y_{1m}Y_{3p})g_i + (Y_{1m}Y_3 - Y_1Y_{3m})g_{i+1}}{\Omega} \\ c &= \frac{(Y_{1p}Y_2 - Y_1Y_{2p})g_{i-1} + (Y_{1m}Y_{2p} - Y_{1p}Y_{2m})g_i + (Y_1Y_{2m} - Y_{1m}Y_2)g_{i+1}}{\Omega}, \end{aligned} \tag{B.5}$$

where $\Omega = Y_{1p}Y_2Y_{3m} - Y_1Y_{2p}Y_{3m} - Y_{1m}Y_2Y_{3p} + Y_1Y_{2m}Y_{3p} + Y_{1m}Y_{2p}Y_3 - Y_{1p}Y_{2m}Y_3$. The distribution of mass density within bin x_i is now given by Equation B.2 and the distribution of mass density within bin x_j is calculated similarly. The two mass density functions of the bins containing the colliding droplets are:

$$m(y_i) = ay_i^2 + by_i + c \quad (\text{B.6a})$$

$$m(y_j) = dy_j^2 + ey_j + f, \quad (\text{B.6b})$$

where d, e , and f are the coefficients of the mass density function of bin x_j and are calculated similarly.

Linear Reconstruction

For the special cases where $i = 1$ or $i = n$, there is only one bin adjacent to either the first or last bins. Since the quadratic interpolation required two adjacent bins, a linear interpolation scheme is used for these bins. This linear interpolation scheme is implemented by solving a system of two equations and two unknowns. The linear scheme applied to the first bin is shown here. A linear interpolation for the last bin is done analogously.

Assume the first two bins have the same intra-bin distribution, $m_l(y)$, and use the following two density formulas, for bin 1 and bin 2 (i.e. $i = 1$ and $i = 2$), respectively.

$$\int_{y_{1-\frac{1}{2}}}^{y_{1+\frac{1}{2}}} m_l(\hat{y}) d\hat{y} = g_1, \quad \int_{y_{2-\frac{1}{2}}}^{y_{2+\frac{1}{2}}} m_l(\hat{y}) d\hat{y} = g_2, \quad (\text{B.7})$$

where $m_l(y)$ is a linear form of the mass density function within bin 1:

$$m_{l,1}(y) = p_1y + q_1. \quad (\text{B.8})$$

Integrating the linear polynomial Equation B.8, in each of the two formulas in Equation B.7, gives a system of two equations in two unknowns:

$$\begin{aligned} \frac{1}{2}(y_{1+\frac{1}{2}}^2 - y_{1-\frac{1}{2}}^2)p_1 + (y_{1+\frac{1}{2}} - y_{1-\frac{1}{2}})q_1 &= g_1 \\ \frac{1}{2}(y_{2+\frac{1}{2}}^2 - y_{2-\frac{1}{2}}^2)p_1 + (y_{2+\frac{1}{2}} - y_{2-\frac{1}{2}})q_1 &= g_2. \end{aligned} \quad (\text{B.9})$$

Solving (B.9) for p_1 and q_1 gives the coefficients for the linear form of the mass density function, (B.8):

$$\begin{aligned} p_1 &= \frac{g_2 \hat{Y}_{12}}{\hat{Y}_{12} \hat{Y}_{21} - \hat{Y}_{11} \hat{Y}_{22}} \\ q_1 &= \frac{g_1 \hat{Y}_{21}}{\hat{Y}_{12} \hat{Y}_{21} - \hat{Y}_{11} \hat{Y}_{22}}, \end{aligned} \quad (\text{B.10})$$

where $\hat{Y}_{11} = \frac{1}{2}(y_{1\frac{1}{2}}^2 - y_{\frac{1}{2}}^2)$, $\hat{Y}_{12} = y_{1\frac{1}{2}} - y_{\frac{1}{2}}$, $\hat{Y}_{21} = \frac{1}{2}(y_{2\frac{1}{2}}^2 - y_{1\frac{1}{2}}^2)$, and $\hat{Y}_{22} = y_{2\frac{1}{2}} - y_{1\frac{1}{2}}$.

The linear interpolation for the last bin, bin n , is

$$m_{l,n}(y) = p_n y + q_n, \quad (\text{B.11})$$

with the coefficients for the linear form of the mass density function, (B.11):

$$\begin{aligned} p_n &= \frac{g_2 \tilde{Y}_{12}}{\tilde{Y}_{12} \tilde{Y}_{21} - \tilde{Y}_{11} \tilde{Y}_{22}} \\ q_n &= \frac{g_1 \tilde{Y}_{21}}{\tilde{Y}_{12} \tilde{Y}_{21} - \tilde{Y}_{11} \tilde{Y}_{22}}, \end{aligned} \quad (\text{B.12})$$

where $\tilde{Y}_{11} = \frac{1}{2}(y_{n+\frac{1}{2}}^2 - y_{n-\frac{1}{2}}^2)$, $\tilde{Y}_{12} = y_{n+\frac{1}{2}} - y_{n-\frac{1}{2}}$, $\tilde{Y}_{21} = \frac{1}{2}(y_{n-\frac{1}{2}}^2 - y_{n-1\frac{1}{2}}^2)$, and $\tilde{Y}_{22} = y_{n-\frac{1}{2}} - y_{n-1\frac{1}{2}}$.

Alternate piece-wise reconstruction

The primary reconstruction polynomials Equation B.6a, B.6b, B.8, and B.11 gives a pdf of the mass density of bin x_i and x_j . When the polynomial reconstruction gives negative values in a source bin, the reconstruction is not physical and an alternate reconstruction must be used. A cartoon of such an example is shown in Figure B.3. For those times, a piece-wise linear polynomial is derived from the root(s) of the original (quadratic or linear) polynomial. We present a methodology to determine if the original polynomial is valid and to derive an alternate polynomial when the initial reconstruction has failed.

For clarity the discussion concerning an alternative reconstruction polynomial will reference bin x_i . Reconstruction in bin x_j is analogous. The alternative reconstruction within the smallest and largest bins, x_1 and x_n , are

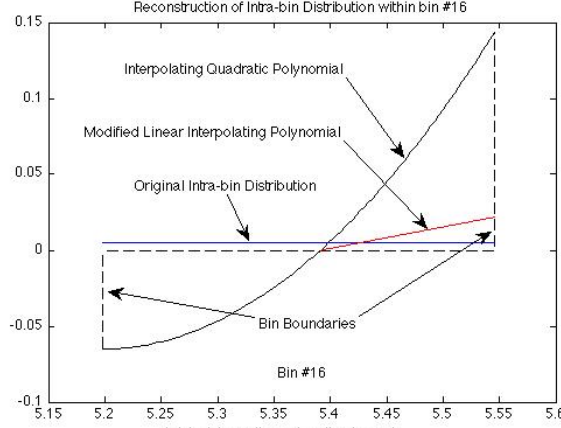
Figure B.3: Source Bins: Alt. Linear Distribution

Figure B.3: A cartoon example of an alternative linear distribution when the quadratic approximation gives negative values for mass density. Bin #16 refers to the log radius coordinates on the abscissa only. The remaining features are for demonstration purposes.

derived separately. The vertex of the quadratic reconstruction polynomial, Equation B.6a, is

$$y_{cr} = \left(\frac{-b}{2a}, \frac{-b^2}{4a} + c \right). \quad (\text{B.13})$$

If $\frac{-b^2}{4a} + c > 0$ then the parabola has no negative values in bin x_i and the reconstruction is valid. If not then the parabolic reconstruction may have negative values in bin x_i , and further conditions need to be tested. For the remainder of this section we will assume that $\frac{-b^2}{4a} + c \leq 0$. We need to determine if the vertex is (I) between the boundaries of bin x_i , (II) to the left of the left bin boundary ($x_{i-di/2}$), or (III) to the right of the right bin boundary ($x_{i+di/2}$). In either of the latter two cases, we need to determine if any root of the parabola lies within the boundaries of bin i . These three cases are shown as rows in Table (B.1). Detailed derivations are given in Appendix A.3.

When there are two roots, the piece-wise linear distribution has three functions. On the interval between the two roots (y_{r1} and y_{r2}), the mass density is set to zero. On the other two intervals the parabolic segments are replaced with linear ones such that the mass is conserved and the ratio of the mass densities in the neighbouring bins is used to approximate the skewness of

Table B.1: Alternate Distribution: Conditions

Conditions Requiring an Alternate Polynomial		
	vertex: ordinate	roots:
I	$\frac{-b^2}{4a} + c \leq 0$	$(y_{r1} > y_{i-di/2})$ and $(y_{r2} < y_{i+di/2})$
II	$\frac{-b^2}{4a} + c \leq 0$	$(y_{r1} < y_{i-di/2})$ and $(y_{r2} < y_{i+di/2})$
III	$\frac{-b^2}{4a} + c \leq 0$	$(y_{r1} > y_{i-di/2})$ and $(y_{r2} > y_{i+di/2})$

Table B.1: Three pieces of information are used to determine when the piece-wise linear reconstruction is used: (i) sign of the ordinate of the vertex, (ii) the location of the first root relative to the left boundary of bin x_i , and (iii) the location of the second root relative to the right boundary of bin x_i .

the distribution. The two linear segments take the form:

$$m_1(y_i) = \alpha(y_{r1} - y) \quad \text{and} \quad m_2(y_i) = \beta(y - y_{r2}), \quad \text{respectively.} \quad (\text{B.14})$$

To conserve mass, the sum of the area under these two segments is set to equal the total mass density in the bin g_i .

$$\int_{y_{i-\frac{di}{2}}}^{y_{r1}} \alpha(y_{r1} - y) dy + \int_{y_{r2}}^{y_{i+\frac{di}{2}}} \beta(y - y_{r2}) dy = g_i. \quad (\text{B.15})$$

We use the ratio of the mass densities in the neighbouring bins to preserve the skewness, and the following equation preserves the ratio of masses that are in the adjacent bins:

$$g_{i+1} \int_{y_{i-\frac{di}{2}}}^{y_{r1}} \alpha(y_{r1} - y) dy = g_{i-1} \int_{y_{r2}}^{y_{i+\frac{di}{2}}} \beta(y - y_{r2}) dy \quad (\text{B.16})$$

We determine the 3-function form of the piece-wise linear distribution by solving (B.15) and (B.16) for α and β :

$$\alpha = \frac{-g_i g_{i-1}}{\Omega_1(g_{i+1} - g_{i-1})} \quad \text{and} \quad \beta = \frac{-g_i g_{i-1}}{\Omega_2(g_{i+1} - g_{i-1})}, \quad (\text{B.17})$$

where $\Omega_1 = \frac{1}{2}y_{r1}^2 - y_{r1}y_{i-\frac{di}{2}} + \frac{1}{2}y_{i-\frac{di}{2}}^2$ and $\Omega_2 = \frac{1}{2}y_{r2}^2 - y_{r2}y_{i+\frac{di}{2}} + \frac{1}{2}y_{i+\frac{di}{2}}^2$. Corresponding to row I of Table B.1, the 3-function piece-wise linear pdf of the mass density function in bin x_i is

$$m_{1,2}(y_i) = \begin{cases} \alpha(y_{r1} - y_i) & \text{if } y_i \in (y_{i-\frac{di}{2}}, y_{r1}) \\ 0 & \text{if } y_i \in (y_{r1}, y_{r2}) \\ \beta(y_i - y_{r2}) & \text{if } y_i \in (y_{r2}, y_{i+\frac{di}{2}}). \end{cases} \quad (\text{B.18})$$

When there is one root of the reconstruction polynomial, the piece-wise linear distribution has two functions. Again, the mass density is set to zero on the interval where the parabola is negative. For two-component piece-wise reconstructions, the same alternate algorithm is used whether the original reconstruction is quadratic or linear. In rows II or III of Table B.1, this interval is to the left or the right of the single root, respectively. On the other interval, the pdf of the mass density is represented by a single, non-zero, linear function. The location of the root corresponding to row II of Table B.1 is shown in Figure B.3. The area under the first curve in (B.14) is set equal to g_i , and solved for the single parameter, α .

$$\int_{y_{i-\frac{di}{2}}}^{y_{r1}} \alpha(y_{r1} - y)dy = g_i \quad \text{which gives} \quad \alpha = \frac{g_i}{\Omega_1}. \quad (\text{B.19})$$

Corresponding to row III of Table B.1, the 2-function piece-wise linear pdf of the mass density function in bin x_i is

$$m_{1*}(y_i) = \begin{cases} \alpha(y_{r1} - y_i) & \text{if } y_i \in (y_{i-\frac{di}{2}}, y_{r1}) \\ 0 & \text{if } y_i \in (y_{r1}, y_{i+\frac{di}{2}}). \end{cases} \quad (\text{B.20})$$

Similarly, for the vertex location given in row II of Table B.1, the piece-wise linear pdf of the mass density function in bin x_i with $\beta = \frac{g_i}{\Omega_2}$ is

$$m_{2*}(y_i) = \begin{cases} 0 & \text{if } y_i \in (y_{i-\frac{di}{2}}, y_{r2}) \\ \beta(y_i - y_{r2}) & \text{if } y_i \in (y_{r2}, y_{i+\frac{di}{2}}). \end{cases} \quad (\text{B.21})$$

The values of the inter-bin distribution of mass density are used to construct a linear approximation of the intra-bin distribution. The three alternate

piece-wise reconstructions correspond to the three rows in Table (B.1), respectively.

The reconstruction polynomials for the source bins are given by Equations B.6a, B.6b, B.8, B.11, B.18, B.20, or B.21.

B.1.2 Moments of Source Bin Distributions

The moments of the probability distribution of the reconstruction polynomial are computed in physical coordinates, and $y = \ln x$ implies $x(y) = \exp(y)$. The zeroth moment is simply the mass contained in each bin. The first moment of the redistribution polynomials are computed using:

$$\overline{x_i} = \frac{1}{g_i} \int_{y_i - \frac{d_i}{2}}^{y_i + \frac{d_i}{2}} x(y) m_i(y) dy = \frac{1}{g_i} \int_{y_i - \frac{d_i}{2}}^{y_i + \frac{d_i}{2}} \exp(y) m_i(y) dy, \quad (\text{B.22})$$

and the second moments are computed with:

$$\overline{x_i^2} = \frac{1}{g_i} \int_{y_i - \frac{d_i}{2}}^{y_i + \frac{d_i}{2}} x^2(y) m_i(y) dy = \frac{1}{g_i} \int_{y_i - \frac{d_i}{2}}^{y_i + \frac{d_i}{2}} \exp(2y) m_i(y) dy. \quad (\text{B.23})$$

The various forms of the reconstruction polynomial given by $m_i(y)$ in Equations B.6a, B.6b, B.8, B.11, B.18, B.20, or B.21 are used to compute the first two moments. The first moments for these six forms are listed in Table A.6 and A.7. The second moments for these six forms are listed in Table A.8 and A.9.

Joint Distribution

The zeroth moment of the joint probability distribution is the sum of the pre collision masses and is given by Equation 2.14. The first moment $\overline{x_{ij}^*}$ of the joint intra-bin distribution of the post collision droplets is derived as follows:

$$\overline{x_{ij}^*} = \frac{1}{\dot{g}_{ij}} \int_{y_i - \frac{d_i}{2}}^{y_i + \frac{d_i}{2}} \int_{y_j - \frac{d_j}{2}}^{y_j + \frac{d_j}{2}} (\exp(\hat{y}_i) + \exp(\hat{y}_j)) m(\hat{y}_i) m(\hat{y}_j) d\hat{y}_i d\hat{y}_j \quad (\text{B.24})$$

where

$$\dot{g}_{ij} = \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} m(\hat{y}_i) m(\hat{y}_j) d\hat{y}_i d\hat{y}_j = \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} m(\hat{y}_i) d\hat{y}_i \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} m(\hat{y}_j) d\hat{y}_j = g_i g_j.$$

since

$$g_i \equiv \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} m(\hat{y}_i) d\hat{y}_i \quad \text{and} \quad g_j \equiv \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} m(\hat{y}_j) d\hat{y}_j.$$

Assuming independence between the two Equations B.6a and B.6b, then Equation B.24 is readily solved.

$$\begin{aligned} \overline{x_{ij}^*} &= \frac{1}{\dot{g}_{ij}} \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} \exp(\hat{y}_i) m(\hat{y}_i) d\hat{y}_i \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} m(\hat{y}_j) d\hat{y}_j + \\ &\quad \frac{1}{\dot{g}_{ij}} \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} m(\hat{y}_i) d\hat{y}_i \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} \exp(\hat{y}_j) m(\hat{y}_j) d\hat{y}_j \\ \overline{x_{ij}^*} &= \frac{1}{g_i} \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} \exp(\hat{y}_i) m(\hat{y}_i) d\hat{y}_i + \frac{1}{g_j} \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} \exp(\hat{y}_j) m(\hat{y}_j) d\hat{y}_j = \overline{x_i^*} + \overline{x_j^*} \end{aligned}$$

Thus $\overline{x_{ij}^*} = \overline{x_i^*} + \overline{x_j^*}.$ (B.25)

The 2^{nd} moment of the joint intra-bin distribution of the mass density of the post collision droplets is derived from the first and second moments of the distributions mass densities of the source bins:

$$\overline{x_{ij}^2} = \frac{1}{\dot{g}_{ij}} \int_{y_{i-\frac{d_i}{2}}}^{y_{i+\frac{d_i}{2}}} \int_{y_{j-\frac{d_j}{2}}}^{y_{j+\frac{d_j}{2}}} (\exp(\hat{y}_i) + \exp(\hat{y}_j))^2 m(\hat{y}_i) m(\hat{y}_j) d\hat{y}_i d\hat{y}_j$$

$$\begin{aligned}
\overline{x_{ij}^2} = & \frac{1}{g_i g_j} \int_{y_{i-\frac{d_i}{2}}^{y_{i+\frac{d_i}{2}}}} \exp(\hat{y}_i)^2 m(\hat{y}_i) d\hat{y}_i \int_{y_{j-\frac{d_j}{2}}^{y_{j+\frac{d_j}{2}}}} m(\hat{y}_j) d\hat{y}_j + \\
& \frac{2}{g_i g_j} \int_{y_{i-\frac{d_i}{2}}^{y_{i+\frac{d_i}{2}}}} \exp(\hat{y}_i) m(\hat{y}_i) d\hat{y}_i \int_{y_{j-\frac{d_j}{2}}^{y_{j+\frac{d_j}{2}}}} \exp(\hat{y}_j) m(\hat{y}_j) d\hat{y}_j + \\
& \frac{1}{g_i g_j} \int_{y_{i-\frac{d_i}{2}}^{y_{i+\frac{d_i}{2}}}} m(\hat{y}_i) d\hat{y}_i \int_{y_{j-\frac{d_j}{2}}^{y_{j+\frac{d_j}{2}}}} \exp(\hat{y}_j)^2 m(\hat{y}_j) d\hat{y}_j.
\end{aligned}$$

For the polynomial reconstruction, the 2^{nd} moment is

$$\begin{aligned}
\overline{x_{ij}^2} = & \frac{1}{g_i} \int_{y_{i-\frac{d_i}{2}}^{y_{i+\frac{d_i}{2}}}} \exp(\hat{y}_i)^2 (a\hat{y}_i^2 + b\hat{y}_i + c) d\hat{y}_i + 2\overline{x_i^*} \overline{x_j^*} + \\
& \frac{1}{g_j} \int_{y_{j-\frac{d_j}{2}}^{y_{j+\frac{d_j}{2}}}} \exp(\hat{y}_j)^2 (d\hat{y}_j^2 + e\hat{y}_j + f) d\hat{y}_j \\
= & 2\overline{x_i^*} \overline{x_j^*} + \overline{x_i^2} + \overline{x_j^2}. \tag{B.26}
\end{aligned}$$

These three moments of the post-collision mass density distribution function are solely dependant on known quantities that are related to the bins of the pre-collision droplets, and are used to form the pdf of the post-collision droplets in the redistribution algorithm.

B.1.3 Target Bin Distribution

The probability distribution of the post collision mass is also computed directly from an assumed distribution in the target bins. Let $m_a(y)$ be the mass density function of the post collision droplets. The first three moments

of the joint density function are given by

$$1 = \frac{1}{\dot{g}_k} \int_{l_{ij}}^{u_{ij}} m_a(\hat{y}) d\hat{y}, \quad \overline{x_{ij}^*} = \int_{l_{ij}}^{u_{ij}} \exp(\hat{y}) m_a(\hat{y}) d\hat{y},$$

$$\overline{x_{ij}^2} = \int_{l_{ij}}^{u_{ij}} \exp(2\hat{y}) m_a(\hat{y}) d\hat{y}, \quad (\text{B.27})$$

$$\text{where} \quad m_a(y) = \alpha y^2 + \beta y + \gamma, \quad (\text{B.28})$$

and (l_{ij}, u_{ij}) is the interval into which the post-collision mass is placed. This interval does not necessarily coincide with the bin boundaries on the log-radius grid (Wang et al. 2007). We calculate the target interval (l_{ij}, u_{ij}) from source bin information. If the distribution of the pre-collision mass spans the entire bin, then the bounds of the bin are used. In this case the bounds of source bins x_i and x_j are $(y_{i-\frac{d_i}{2}}, y_{i+\frac{d_i}{2}})$ and $(y_{j-\frac{d_j}{2}}, y_{j+\frac{d_j}{2}})$, respectively and $(l_{ij}, u_{ij}) = (y_{i+j-\frac{d_i+d_j}{2}}, y_{i+j+\frac{d_i+d_j}{2}})$. In an alternate reconstruction, the roots of the quadratic polynomial may be substituted for the bounds of the bin. If for example, Equation B.20 is the distribution function for the pre-collision mass of bin i , then $(l_{ij}, u_{ij}) = (y_{i+j-\frac{d_i+d_j}{2}}, y_{r1} + y_{j+\frac{d_j}{2}})$. If Equation B.20 is the distribution function for the pre-collision mass of bin i and Equation B.21 is the distribution function for the pre-collision mass of bin j , then $(l_{ij}, u_{ij}) = (y_{i-\frac{d_i}{2}} + y_{r2}, y_{r1} + y_{j+\frac{d_j}{2}})$.

Substituting Equation B.28 into Equation B.27 and having already computed the values of the left hand sides of the three Equations B.27, we get three equations with three unknowns:

$$1 = \int_{l_{ij}}^{u_{ij}} (\alpha \hat{y}^2 + \beta \hat{y} + \gamma) d\hat{y},$$

$$\overline{x_{ij}^*} = \int_{l_{ij}}^{u_{ij}} \exp(\hat{y}) (\alpha \hat{y}^2 + \beta \hat{y} + \gamma) d\hat{y}, \quad (\text{B.29})$$

$$\overline{x_{ij}^2} = \int_{l_{ij}}^{u_{ij}} \exp(2\hat{y}) (\alpha \hat{y}^2 + \beta \hat{y} + \gamma) d\hat{y}.$$

The integrations of the functions Equation B.29 are easily computed:

$$\begin{aligned}
1 &= \frac{1}{3}(u_{ij}^3 - l_{ij}^3)\alpha + \frac{1}{2\dot{g}_{ij}}(u_{ij}^2 - l_{ij}^2)\beta + \frac{1}{\dot{g}_{ij}}(u_{ij} - l_{ij})\gamma, \\
\overline{x_{ij}^*} &= \alpha \left[(2 - 2y_{i+di/2} + y_{i+di/2}^2) \exp(y_{i+di/2}) - \right. \\
&\quad \left. (2 - 2y_{i-di/2} + y_{i-di/2}^2) \exp(y_{i-di/2}) \right] + \quad (B.30) \\
&\quad \beta \left[(y_{i+di/2} - 1) \exp(y_{i+di/2}) - (y_{i-di/2} - 1) \exp(y_{i-di/2}) \right] + \\
&\quad \gamma \left[\exp(y_{i+di/2}) - \exp(y_{i-di/2}) \right], \\
\overline{x_{ij}^2} &= \frac{\alpha}{4} \left[(1 - 2y_{i+di/2} + 2y_{i+di/2}^2) \exp(2y_{i+di/2}) - \right. \\
&\quad \left. (1 - 2y_{i-di/2} + 2y_{i-di/2}^2) \exp(2y_{i-di/2}) \right] + \\
&\quad \frac{\beta}{2} \left[(y_{i+di/2} - 0.5) \exp(2y_{i+di/2}) - (y_{i-di/2} - 0.5) \exp(2y_{i-di/2}) \right] + \\
&\quad \frac{\gamma}{2} \left[\exp(2y_{i+di/2}) - \exp(2y_{i-di/2}) \right]. \quad (B.31)
\end{aligned}$$

The Equations B.30 and B.31 are linear in α , β , and γ , and are solved in a similar manner as the 3×3 system in Section B.1.1. Substituting $\overline{Y}_{11}$ for $\frac{1}{3}(u_{ij}^3 - l_{ij}^3)$ and making the eight other corresponding substitutions in Equations B.30 and B.31 simplifies the system of equations to:

$$\begin{aligned}
\overline{Y}_{11}\alpha + \overline{Y}_{12}\beta + \overline{Y}_{13}\gamma &= 1, \\
\overline{Y}_{21}\alpha + \overline{Y}_{11}\beta + \overline{Y}_{12}\gamma &= \overline{y_{ij}^*}, \\
\overline{Y}_{31}\alpha + \overline{Y}_{21}\beta + \overline{Y}_{11}\gamma &= \overline{y_{ij}^2}. \quad (B.32)
\end{aligned}$$

Solving the system of equations (B.32) for α , β , and γ gives the coefficients for the quadratic form of the joint mass density function (B.28):

$$\begin{aligned}
\alpha &= \frac{(Y_{22}Y_{33} - Y_{23}Y_{32}) + (Y_{13}Y_{32} - Y_{12}Y_{33})\overline{y_{ij}^*} + (Y_{12}Y_{23} - Y_{13}Y_{22})\overline{y_{ij}^2}}{\overline{\Omega}}, \\
\beta &= \frac{(Y_{31}Y_{23} - Y_{21}Y_{33}) + (Y_{11}Y_{33} - Y_{31}Y_{13})\overline{y_{ij}^*} + (Y_{21}Y_{13} - Y_{11}Y_{23})\overline{y_{ij}^2}}{\overline{\Omega}}, \\
\gamma &= \frac{(Y_{21}Y_{32} - Y_{31}Y_{22}) + (Y_{31}Y_{12} - Y_{11}Y_{32})\overline{y_{ij}^*} + (Y_{11}Y_{22} - Y_{21}Y_{12})\overline{y_{ij}^2}}{\overline{\Omega}}, \quad (B.33)
\end{aligned}$$

where $\bar{\Omega} = Y_{31}Y_{12}Y_{23} - Y_{31}Y_{13}Y_{22} - Y_{21}Y_{12}Y_{33} + Y_{21}Y_{13}Y_{32} + Y_{11}Y_{22}Y_{33} - Y_{11}Y_{23}Y_{32}$.

The mass density function of the post-collision droplets, (B.28), and the corresponding coefficients in (B.33) are fully described by known quantities related to the pre-collision droplets.

B.1.4 Target Mass Allocation

Figure B.4: Target Bins: Quadratic Distribution

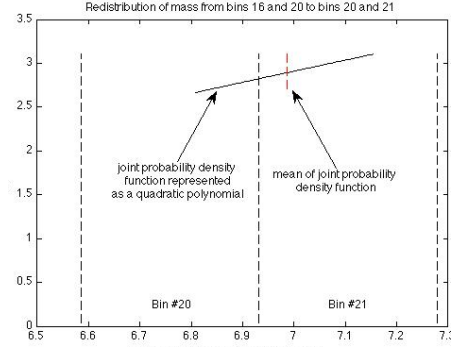


Figure B.4: Target bins x_k and x_{k+1} are shown the coartroom of mass distribution into two target bins. Bins #20 and 21 refer to the log radius coordinates on the abscissa only. The remaining features are for demonstration purposes.

The mass density Equation 2.14 that is created as a result of two colliding droplets is restricted to being placed in the interval (l_{ij}, u_{ij}) . A target bin (x_k) is chosen that contains the first moment of the joint density function: $y_{k-dk/2} \leq \bar{y}_{ij}^* \leq y_{k+dk/2}$. Since $\bar{y}_{ij}^* \in (l_{ij}, u_{ij})$, this interval can cover two of three possible bins: x_{k-1} , x_k , or x_{k+1} . By the left most equation of Equation B.27, some of the parabola takes positive values in the interval (l_{ij}, u_{ij}) . We claim that if the parabola takes positive values anywhere in the interval (l_{ij}, u_{ij}) , then it takes positive values everywhere in that interval. We support this claim by defining the domain in which Reconstruction and Redistribution Algorithm is valid and noting that the reconstructed mass density functions in the source bins take non-negative values over their entire respective domains. The shape, orientation, and location (relative to l_{ij} and u_{ij}) of the parabola determines how the post-collision mass $\acute{g}_{ij}$ is distributed among the three possible bins. An example is shown in Figure B.4. The mass density function Equation B.28 is integrated to determine the quantity

of mass density placed in each target bin.

$$m_{a,n}(y) = \int_{n_{\text{lower}}}^{n_{\text{upper}}} m_a(\hat{y}) d\hat{y}, \quad (\text{B.34})$$

where n_{lower} and n_{upper} are the bound of integration and are specified in Table B.2.

Table B.2: Target Bins: Integration Limits

Figure	x_{k-1}		x_k		x_{k+1}	
	n_{lower}	n_{lower}	n_{lower}	n_{lower}	n_{lower}	n_{lower}
B.4	—	—	l_{ij}	$y_{k+dk/2}$	$y_{k+dk/2}$	u_{ij}
not shown	l_{ij}	$y_{k-dk/2}$	$y_{k-dk/2}$	u_{ij}	—	—
not shown	—	—	l_{ij}	u_{ij}	—	—

Table B.2: The bounds of integration of the joint mass density function for each target bin. The left column identifies the associated Figure for distributions spanning two target bins.

There are three possible arrangements of l_{ij} and u_{ij} relative to $y_{k-dk/2}$, and $y_{k+dk/2}$. When $(l_{ij}, u_{ij}) \in (y_{k-dk/2}, y_{k+dk/2})$ then all the mass density is trivially placed in bin x_k . In Figure B.4, $l_{ij} > y_{k-dk/2}$, and $u_{ij} > y_{k+dk/2}$ and the mass is split between bins x_k and x_{k+1} . The bounds of integration for each of the target bins are detailed in Table B.2. Because of mass conservation, the number of integrations that are necessary are one less than the number of bins into which the mass density is redistributed.

B.1.5 Alternative Target Distribution

The redistribution curve conserves the first three moments of the mass density distribution. When the evolution of the mass distribution function is governed by a collision kernel more complicate than a simple mass-of-masses function, then the redistribution curve can take negative values in the collision domain. An example is shown in Figure B.5.

As with the reconstruction curve, negative values are non-physical and an alternative redistribution curve must be built. The alternative redistribution conserves the mass and centroid of the target mass. The redistribution

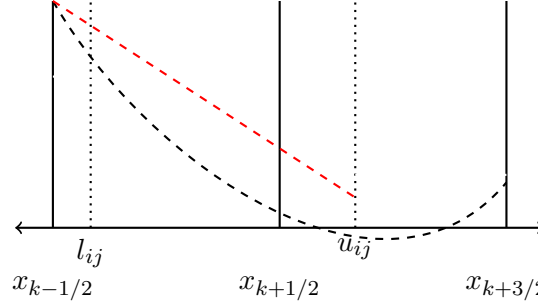
Figure B.5: Target Bins: Negative Mass Density

Figure B.5: A cartoon of a quadratic distribution curve over two target bins is shown as a dotted line in black. The dotted red line is an alternate linear distribution curve created by relaxing the requirement to conserve the variance, and retaining only conservation of mass and conservation of the centroid.

curves in Figure B.5 show negative mass because the variance in the assumed distribution is unnaturally large. A reasonable, and physical, response is to relax the conservation of variance while maintaining conservation of mass g_k and centroid $\bar{x}_{ij}$. We assume a linear distribution of the form

$$p_2(x) = g_l + \frac{g_u - g_l}{u_{ij} - l_{ij}}(x - l_{ij})$$

where $g_l = p(l_{ij})$ and $g_u = p(u_{ij})$.

Figure B.6 gives the possible configurations of the target domain w.r.t the bounds of the target bins.

Conservation of mass of and conservation of the centroid are enforced using

$$g_k = \int_{l_{ij}}^{u_{ij}} p_2(x) dx \quad \text{and} \quad \int_{l_{ij}}^{\bar{x}_{ij}} p_2(x) dx = \int_{\bar{x}_{ij}}^{u_{ij}} p_2(x) dx, \quad \text{respectively.} \quad (\text{B.35})$$

Note that the centroid $\bar{x}_{ij}$ doesn't necessarily equal the midpoint of any target bin x_k . Equation (B.35) is a linear system of two equations and two

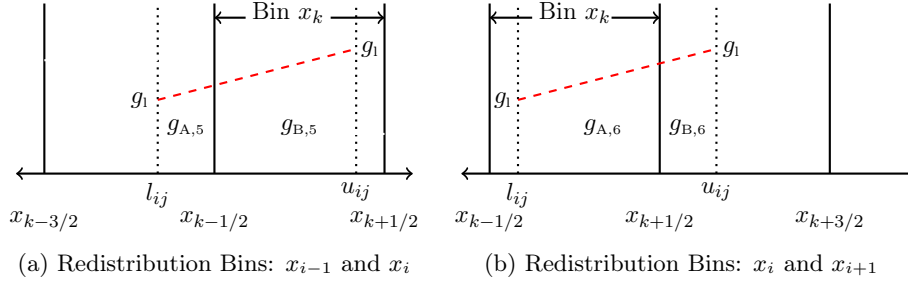
Figure B.6: Target Bins: Alternate Distribution

Figure B.6: The two graphs represent the possible arrangements of the target domain w.r.t. the boundaries of two target bins using the conservative alternative redistribution.

unknowns: g_u and g_l such that:

$$g_l = g_k \frac{2\bar{x}_{ij}^2 - 4\bar{x}_{ij}l_{ij} + l_{ij}^2 - u_{ij}^2 + 2u_{ij}l_{ij}}{2(u_{ij} - l_{ij})(\bar{x}_{ij}^2 - \bar{x}_{ij}^2l_{ij} - l_{ij}^2 + u_{ij}l_{ij} - \bar{x}_{ij}u_{ij})},$$

$$g_u = g_k \frac{2\bar{x}_{ij}^2 - 4\bar{x}_{ij}u_{ij} - 5l_{ij}^2 - u_{ij}^2 + 2u_{ij}l_{ij}}{2(u_{ij} - l_{ij})(\bar{x}_{ij}^2 - \bar{x}_{ij}^2l_{ij} - l_{ij}^2 + u_{ij}l_{ij} - \bar{x}_{ij}u_{ij})}.$$

The masses added to the target bins in the graph on the left in Figure (B.6) are

$$g_{A,5} = (\bar{x}_{k-1/2} - l_{ij}) \left[g_l + \frac{g_u - g_l}{u_{ij} - l_{ij}} \frac{\bar{x}_{k-1/2} - l_{ij}}{2} \right],$$

$$g_{B,5} = (u_{ij} - \bar{x}_{k-1/2}) \left[g_l + \frac{g_u - g_l}{u_{ij} - l_{ij}} \left(\frac{u_{ij} + \bar{x}_{k-1/2}}{2} - l_{ij} \right) \right].$$

The masses added to the target bins in the graph on the right in Figure (B.6) are

$$g_{A,6} = (\bar{x}_{k+1/2} - l_{ij}) \left[g_l + \frac{g_u - g_l}{u_{ij} - l_{ij}} \frac{\bar{x}_{k+1/2} - l_{ij}}{2} \right],$$

$$g_{B,6} = (u_{ij} - \bar{x}_{k+1/2}) \left[g_l + \frac{g_u - g_l}{u_{ij} - l_{ij}} \left(\frac{u_{ij} + \bar{x}_{k+1/2}}{2} - l_{ij} \right) \right].$$

B.1.6 Same-bin Collisions

When $i = j$ the probability density functions, ($m(y_i)$ and $m(y_j)$), of the bins containing the two colliding droplets are not independent, and the recon-

struction algorithm is not valid. In the case of $i = j$ all the mass density is placed in bin x_k where $k = i + s$. When $i = j$, all the mass density from the collisions of droplets from bin x_i is placed into bin x_k . The maximum interval into which the mass density of the post-collision droplets can be placed is twice the width of bin x_i . The length of this maximum interval is shown to be smaller than the width of bin x_k , and therefore, the post collision mass in same bin collisions is placed in a single target bin. Since the width of bin x_i is $(y_{i-di/2}, y_{i+di/2})$, then twice the width of bin x_i is (y_{2i-di}, y_{2i+di}) . The width of bin x_k is $(y_{2i-d2i/2}, y_{2i+d2i/2})$ as shown in Figure B.7.

Figure B.7: Same Bins

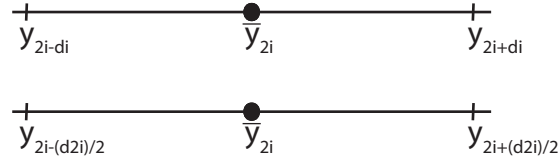


Figure B.7: The edges of bin x_{2i} are represented in two ways. Top: as double the width of bin x_i . Bottom: as the width of bin $2i$ directly.

Successive bin widths are calculated by $x_{n+1} = \alpha x_n$ where $\alpha = 2^{1/z}$ and z is a natural number. The width of the bin doubles every z^{th} bin, so $k = i + z$. Assuming the edges of the bins are halfway between the centers of adjacent bins, the edges of bin n are

$$x_{n+dn/2} = \frac{x_n + x_{n+1}}{2} = \frac{x_n + \alpha x_n}{2} = \frac{\alpha + 1}{2} x_n,$$

and

$$x_{n-dn/2} = \frac{x_{n-1} + x_n}{2} = \frac{x_{n-1} + \alpha x_{n-1}}{2} = \frac{\alpha + 1}{2} x_{n-1}.$$

So, the width of bin x_n is

$$x_{n+dn/2} - x_{n-dn/2} = \frac{\alpha + 1}{2} x_n - \frac{\alpha + 1}{2} x_{n-1} =$$

$$\frac{\alpha + 1}{2} \alpha x_{n-1} - \frac{\alpha + 1}{2} x_{n-1} = \frac{\alpha^2 - 1}{2} x_{n-1}.$$

The width of bin n is taken as a function of the width of the smallest bin, x_0 .

$$d_n = x_{n+dn/2} - x_{n-dn/2} = \frac{\alpha^2 - 1}{2} \alpha^{n-1} x_0. \quad (\text{B.36})$$

By Equation B.36, twice the width of bin i , $d_i = |y_{2i-di}, y_{2i+di}|$, is $2d_i = (\alpha^2 - 1)\alpha^{i-1}x_0$; and the width of bin $2i$, $d_{2i} = |y_{2i-d2i/2}, y_{2i+d2i/2}|$, is $d_{2i} =$

$(\frac{\alpha^2 - 1}{2})\alpha^{2i-1}x_0$. We construct a ratio of the maximum interval of the post-collision mass density to the width of the target bin.

$$\frac{2d_i}{d_{2i}} = \frac{(\alpha^2 - 1)\alpha^{i-1}x_0}{(\frac{\alpha^2 - 1}{2})\alpha^{2i-1}x_0} = \frac{2\alpha^{i-1}}{\alpha^{2i-1}} = \frac{2}{\alpha^i}. \quad (\text{B.37})$$

If $\alpha \geq 2$ and $i \geq 1$ or $\alpha \geq \sqrt[3]{2}$ and $i \geq p$, for $n \geq 2$, then $|2di| \leq |d2i|$. Given Equation B.25, $2di \subseteq d2i$, and therefore all the mass density from the collision of droplets in the same bin, x_i , will be contained in a bin that is $s \geq i \geq 1$ times the width of bin x_i . So, no mass density is fluxed between multiple target bins when $i = j$.

Table B.3: Computational Expense

Method	Intra-Bin Distribution	Addition	Multiplication	Exponential
Linear Flux	Constant	11	12	0
Bin Integral	1 st order	28	56	0
Quadratic	2 nd order	91	121	4

Table B.3: The bounds of integration of the joint mass density function for each target bin. The left column identifies the associated Figure for distributions spanning two target bins.

B.1.7 Computational Efficiency

The computational cost of this new detailed bin interaction method is compared against the computationally efficient and popular Linear Flux Method (LFM) (Bott 1998) and a more accurate but also more expensive Bin Integral Method (BIM)(Wang et al. 2007). The BIM constructs an linear intra-bin distribution of mass density, in contrast to the LFM which constructs a constant intra-bin distribution of mass density, and the Reconstruction & Redistribution Algorithm (RRA) which constructs a quadratic intra-bin distribution of mass density. Both the BIM and RRA have multiple algorithms for the distribution intra-bin mass density, because the main algorithm of each method can have a root of the distribution in the source bin.

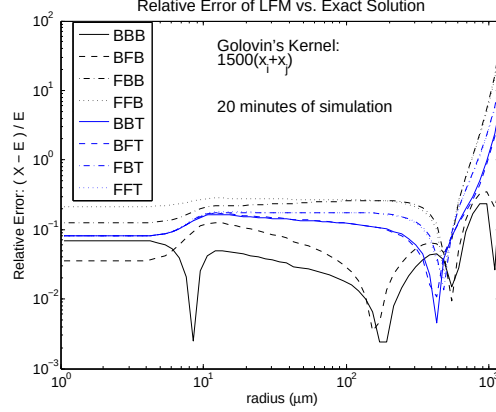
Figure B.8: Collision Sequencing Bias

Figure B.8: The errors of eight sequences of bin-interactions against the analytic solution using Golovin's sum of masses kernel. A 'B' or an 'F' in the first two places in the three letter description refers to the Backward or Forward. A 'B' or an 'T' in the third place refers to Bottom of Top. Further descriptions of the nomenclature are given in Table B.4 and in the text.

For the purposes of the comparison of computational expense, these operations in detailed methods were summed for only the main, or common algorithm. For the BIM that is the centred distribution. For the RRA, it is the quadratic distribution (both source and target bins). These distributions are the more expensive than any of their alternatives. The RRA method using Golovin's sum-of-masses kernel used the quadratic distribution 79.65% of the time during a 60 minute evolution.

The LFM is often used as a benchmark by bulk models (Khairoutdinov & Kogan 2000, Seifert & Beheng 2001, Wang et al. 2007, Franklin 2008). According to Google Scholar at the time of writing this dissertation, the LFM by Bott (1998) was cited 128 times over the last 18 years (three times by the original author), and the BIM by Wang et al. (2007) was cited 15 times over the last nine years (five times by the original authors and twice by the original author's graduate students). Despite being a more accurate method, the BIM has not enjoyed the success and utility that the LFM has. Not counting the original authors' (and graduate students') own citations, the LFM has had nearly eight times the citations that the BIM has. Table B.3 shows that the BIM is approximately 3.6 times as expensive as the LFM. Given the low citation rate of a method that was 3.6 times as expensive as a popular and efficient method, the likelihood of our new method which is

9.4 times as expensive quite hopeful and optimistic at best.

B.1.8 Collision Sequencing Bias

Detailed bin-interaction methods all contain two nested FOR loops that simulate each droplet size interacting with every droplet size. For simplicity assume that the outer loop uses i as the index, the inner one j , and the largest bin is indexed as N . These interactions can be graphically represented in the first quadrant as a triangle bound by $i = j$, $i = 0$, and $j = N$, or by a triangle bound by $i = j$, $j = 0$, and $i = N$. We'll refer to these triangles as the Top and the Bottom, respectively. Within each triangle, the index of the FOR loops can increase or decrease. The eight different possible sequences are given in Table B.4.

Table B.4: Collision Sequence Nomenclature

Abbrev.	Loop Direction		Triangle Half
	Outer: i	Inner: j	
BBB	Backward	Backward	Bottom
BFB	Backward	Forward	Bottom
FBB	Forward	Backward	Bottom
FFB	Forward	Forward	Bottom
BBT	Backward	Backward	Top
BFT	Backward	Forward	Top
FBT	Forward	Backward	Top
FFT	Forward	Forward	Top

Table B.4: The bounds of integration of the joint mass density function for each target bin. The left column identifies the associated Figure for distributions spanning two target bins.

Simulations using Bott's (1998) LFM were performed with each of the eight sequences of bin-interactions. Golovin's sum on masses kernel was used because it is a very simple kernel and the assumption was made that differences would be minimized with a simple kernel and more complex kernels would

likely have greater variation in the relative error. Bott used BBT in his simulations as it was the most accurate of the four in the top triangle, and because it provided an (artificial) brake on the excessive advection of mass towards the larger sized droplets in the droplet spectrum (not shown here).

Figure B.8 shows a consistent order of magnitude difference in the relative error. For some size droplets there is a two order of magnitude difference. The error is not consistent across the droplet spectrum.

B.2 Modified Stochastic Simulation of Collision and Coalescence

Generalizing the bin-pair interaction modification of Gillespie’s stochastic simulation to include positive non-integer droplet numbers is consistent with a ‘quasi-stochastic’ or ‘pure stochastic’ model of coalescence (Gillespie 1975*b*). When a stochastic algorithm uses non-integers to quantify droplet numbers in simulations of a subset of a physical cloud, the evolved droplet size information can be interpreted as the number of drops of each mass or the probability of any number of drops of each mass. In either case by the ergodicity shown by Pope (Wyngaard 2010), an ensemble of simulations of the generalized bin-pair interaction, modified stochastic algorithm gives a mean and variance of droplet masses that can be interpreted to represent a homogeneous portion of a cloud.

These first and second moment statistics can be used to verify the results of detailed spectral methods. A discrete spectral method is, in turn, used to validate bulk parameterizations (Khairoutdinov & Kogan 2000, Seifert & Beheng 2001). Accurate bulk parameterizations are used by cloud resolving models to determine the onset of precipitation and the amount of radar reflectivity (Seifert & Beheng 2001, Franklin 2008). To make this necessary process more widely accessible we present a further modification that eliminates conditions leading to unphysical state spaces.

The discretization process introduces physical requirements on the modified evolution algorithm that were not present when individual droplets were selected by Gillespie’s Monte Carlo Method. The mean mass within a bin is defined (for bin i) by $\bar{x}_i = m_i/N_i$. For the rest of this paper the phrase ‘mean mass’ will refer to the mean mass within a bin. Each collision in a simulation of the modified Gillespie algorithm involves reducing the droplet

number of each selected source bin by unity and choosing, for removal, a droplet mass within the source bin. The droplet mass must be chosen so that the remaining mass and remaining number yields a mean mass that is within the bin boundaries. In Section B.2.1 the droplet selection process of the modified Monte Carlo algorithm by Seeβelberg and collaborators is briefly reviewed. In Section B.2.2 the existence and prevalence of conditions leading to unphysical state spaces are presented, and an example is given. Section B.2.3 details the modification to the droplet selection process used to eliminate unphysical conditions.

B.2.1 Current Droplet Selection Process

The algorithm by Seeβelberg et al. includes a process to select a subinterval within the bin from which to choose a droplet mass. The subinterval is given in their paper as Equation (9). Seeβelberg et al. assumed a uniform distribution of mass density within this subinterval and randomly selected a droplet. In Sections B.2.2 and B.2.3 we maintain that assumption. The interval I_i is constructed to be symmetric about the mean mass $\bar{x}_i$. The masses of droplets at the left and right-hand bin boundaries are denoted as $x_{i-1/2}$ and $x_{i+1/2}$, respectively. Letting $\Delta m_i \equiv \min(x_{i+1/2} - \bar{x}_i, \bar{x}_i - x_{i-1/2})$, the bounds of I_i are defined by (Seesselberg et al. 1996)

$$d_{i-1/2} = \bar{x}_i - \Delta m_i, \quad \text{and} \quad d_{i+1/2} = \Delta m_i + \bar{x}_i,$$

$$I_i = [d_{i-1/2}, d_{i+1/2}] \subseteq [x_{i-1/2}, x_{i+1/2}].$$

B.2.2 Unphysical Conditions

The existence if the conditions that facilitate the unphysical state space is discussed. An example which arises from the initial distribution used by Seeβelberg et al. is detailed, and the prevalence of the condition is noted.

Existence

The droplet selection process given by Seeβelberg et al. fails to adequately restrict the interval I_i when $1 < N_i < 2$. An example of the conditions predicating this problem are present in the initial droplet size distribution used by Seeβelberg et al., and commonly used by others (Berry 1967, Berry &

Reinhardt 1974, Bott 1998, Franklin 2008). This initial gamma distribution is given in the paper by Seeβelberg et al. as their Equation (19). When for example bin i containing $N_i \in (1, 2)$ droplets is selected as the bin containing a pre-collision droplet for the next collision, the removal of too much or too little mass is possible and would result in a post-collision mean mass for bin i that is either less than the mass of the smaller bin boundary, or greater than mass of the larger bin boundary, respectively.

The sketch in Figure (B.9) illustrates conditions that can lead to a mean mass outside of the bin after the collision. The spectrum is described by the mass of a droplet where $\hat{g}_i$ is the total mass in bin i . The two larger (red) arcs are the bounds of interval I_i . A sub-interval bounded by $\hat{g}_i$ and $x_{i+1/2}$ lies within the interval bounded by the larger (red) arcs. Therefore, when N_i is slightly greater than unity, say $N_i = 1 + \alpha$ it is, (i) possible to select an amount of mass to be removed that is greater than the total amount of mass in bin i , and (ii) possible to select an amount of mass that is less than $\hat{g}_i$ but will result in the post-collision mean mass less than $x_{i-1/2}$ as further detailed in the following example.

Figure B.9: Current Droplet Selection Interval

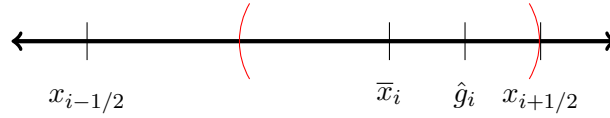


Figure B.9: A schematic of one bin on the droplet size spectrum showing the bin boundaries for bin i , mean intra-bin mass $\bar{x}_i$, and the aggregate mass contained in bin i , $\hat{g}_i$. The larger (red) arcs are the boundaries of the interval within which a droplet is chosen for the next collision. The algorithm to determine these boundaries are by Seeβelberg et al. (1996) and briefly explained in Section B.2.1.

Example

The initial conditions presented in Seeβelberg et al. create conditions in bin #17 of their spectral discretization scheme that potentially lead to a post collision mean mass outside of the bin boundaries even when the mass removed is restricted to an amount less than the mass present. The scaling of the initial distribution by 1500 as reported in their article only serves to shift the bin with $1 < N_j < 2$ downstream. Bin #17 of their unscaled distribution contains 2.046×10^{-08} g and 1.089 droplets. When 1.893×10^{-08} g are removed

then the remaining mass $x_{17,r}$ and remaining droplet number 0.089 gives a mean mass equal to the mass of the smaller bin boundary. Therefore, when a droplet of mass $x_{17,r}$ is removed from bin #17 and $x_{17,r} > 1.893 \times 10^{-08}$ g, the post collision mean mass will be greater than the upper bound of the bin #17.

This flaw can be expressed as $\hat{x}_i < x_{i,r} < \hat{g}_i$ where $\hat{x}_i$ is the maximum mass that can be removed while keeping the post collision mean mass inside of the bin. Having a mean mass outside of a bin that contains a fraction of a droplet such as 0.089 may not seem critical, but the addition of a droplet to bin i , due to a collision with a smaller droplet, will not guarantee that the mean mass will return to the inside of the bin.

Figure B.10: Proposed Droplet Selection Interval

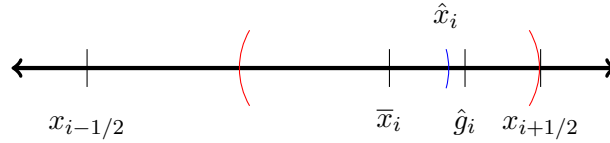


Figure B.10: A schematic of one bin on the droplet size spectrum showing the same as in Figure (B.9) with the addition of a smaller (blue) arc $\hat{x}_i$ that represents the upper bound of a more restrict interval. The calculations used to create this more restrictive boundary are described in the surrounding text.

Prevalence

The prevalent use of a gamma function as an initial distribution requires that bins with $1 < N_i < 2$ will typically be present. The persistence of a practical maximum droplet size (whether through precipitation or breakup) guarantees the existence of the condition in some bin at each time step. A distribution which develops a bimodal character can have multiple bin with potential fatalities. The stochastic nature of the algorithm facilitates a random selection of a source bin, and the possibility that a bin with a critically low droplet number be chosen.

Several simulations have been run without the refinement presented in this dissertation, and many have produced a mean mass external to the bin. These external mean mass occurrences have occurred in multiple bins and at various time steps. Successfully completing a simulation without a fatal error is possible. However, such results may be produced after the emergence

of an unphysical state space at some time step during the simulation which may call into question the validity of the simulation.

B.2.3 A Refinement to the Droplet Selection Process

A sub-interval $\hat{I}_i$ is constructed that is dependent on the bin boundaries ($x_{i-1/2}$ and $x_{i+1/2}$): $\hat{I}_i \subseteq I_i$ such that after the simulation of a collision is complete the mean mass $\bar{x}_i$ will remain between the bin boundaries. The upper bound of $\hat{I}_i$ is shown in Figure (B.10) and subsequently developed herein.

The smaller (blue) arc represents the maximum amount of mass that can be removed such that $\hat{g}_i - \hat{x}_i$ is the amount of mass remaining after the next collision involving bin i and $(\hat{g}_i - \hat{x}_i)/\alpha = \bar{x}_{i,a} = x_{i-1/2}$ where $\bar{x}_{i,a}$ is the mean mass after the collision. The following procedure is used to calculate $\hat{x}_i$ as a function of $x_{i-1/2}$ and also to calculate a more restrictive lower bound as a function of $x_{i+1/2}$.

Figure B.11: Unbiased Proposed Droplet Selection Interval

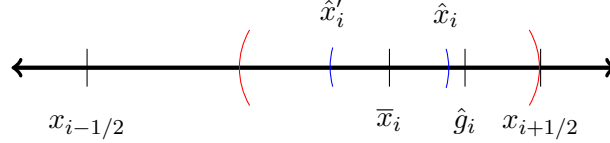


Figure B.11: A schematic of one bin on the droplet size spectrum showing the same as in Figure (B.10) with the addition of a second smaller (blue) arc $\hat{x}'_i$ that represents the lower bound of a more the restrict interval. The calculations used to create the lower bound of this more restrictive interval are in the same spirit as that originally used by Seeßelberg et al. (1996).

The following procedure will give one or more bounds of the more restrictive interval that ensures the remaining mean intra-bin mass will be within the bin boundaries:

$$\begin{aligned}
 e_{i-1/2} &= L_i - x_{i+1/2}(N_i - 1) & e_{i+1/2} &= \hat{x}_i = L_i - x_{i-1/2}(N_i - 1), \\
 \bar{d}_{i-1/2} &= \max(d_{i-1/2}, e_{i-1/2}), & \bar{d}_{i+1/2} &= \min(d_{i+1/2}, e_{i+1/2}), \\
 \bar{I}_i &= [\bar{d}_{i-1/2}, \bar{d}_{i+1/2}] \subseteq I_i \subseteq [x_{i-1/2}, x_{i+1/2}].
 \end{aligned}$$

L_i and N_i are the liquid water content and number concentration in bin i prior to the next collision involving bin i .

In the example from Section B.2.2, $\bar{d}_{i+1/2}$ is a function of the lower bin boundary and is shown in Figure (B.11).

Notice that the interval bounded by the left-most of the larger (red) arcs and $\hat{x}_i$ is not symmetric about $\bar{x}_i$. An interval symmetric about $\bar{x}_i$ is needed to avoid bias. Therefore, a final step in the spirit of the algorithm by Seeβelberg et al. (1996) is used to produce the sub-interval $\hat{I}_i$ bounded by the two smaller (blue) arcs:

$$\hat{x}'_i = \bar{x}_i - (\hat{x}_i - \bar{x}_i)$$

We now have an interval $\hat{I}_i = [\hat{x}'_i, \hat{x}_i]$, symmetric about $\bar{x}_i$, from which to draw the pre-collision droplet which guarantees that the remaining mass and remaining number concentration will yield a post-collision mean intra-bin mass that is inside of the bin boundaries:

$$\hat{I}_i = [\hat{x}'_i, \hat{x}_i] \subseteq \bar{I}_i \subseteq I_i \subseteq [x_{i-1/2}, x_{i+1/2}]$$

B.2.4 Summary

The discretization of the droplet spectrum by Seeβelberg (1996) and collaborators reduced the computational expense of stochastically simulating the collision and coalescence process over the expense of a stochastic simulation that records information about every droplet as given by Gillespie (1975). That modification produce the additional requirement of ensuring that the ratio of intra-bin mass and intra-bin number lie between the two values on the edges of the bin. This requirement is intrinsically met by the algorithm presented by Seeβelberg et al. except for the cases when the post-collision intra-bin number is $0 < N_i < 1$. They restrict the interval in the source bins for the purpose of eliminating droplet selection bias. However, their selection interval does not guarantee that the post-collision mean mass lies within the bin. Situations resulting in such a post collision number concentration has been shown to occur in the commonly used gamma initial distribution, and the condition will persist as the mode traverses the droplet spectrum.

These situations are addressed by further restricting the intra-bin interval contained in the source bins upon which is constructed a uniform distribution. The additional restrictions on the domain from which the mass of a source droplet can be selected are functions of bin boundaries. The new domain is then made symmetric about the intra-bin mean mass to eliminate

selection bias. The restricted intra-bin domain allows for a generalization of initial conditions that include continuous (i.e. non-integer) values of number concentration. There is no need to modify an initial distribution that contains non-integer values of number concentration. Nor is there a need to prevent collisions of droplets in bins containing a number between 1 and 2.