Monte carlo simulations of drop growth by coalescence and collision-induced breakup

L. Alfonso
Universidad Autónoma de la Ciudad de México,
México City, 09790 México,
e-mail: lesterson@yahoo.com

G.B. Raga and D. Baumgardner
Centro de Ciencias de la Atmósfera, Universidad Nacional Autónoma de México,
México City, 04510, México.

Recibido el 7 de mayo de 2009; aceptado el 29 de septiembre de 2009

A Monte Carlo framework to simulate the evolution of drop spectra by coalescence and collision-induced breakup is presented. The stochastic algorithm of Gillespie [1] for chemical reactions in the formulation proposed by Laurenzi and Diamond [2] was used to simulate the kinetic behavior of the drop population. Within Gillespie's framework, the collision-induced breakup process is modeled as a new "chemical reaction". The results of the Monte Carlo simulations were compared with the analytical solution to the collection-breakup equation obtained by Feingold *et al.* [3], for an exponential distribution of satellite drops, and a constant collection and breakup kernels. A good correspondence between the analytical and the stochastic algorithm was found for this case.

Keywords: Cloud microphysics; Monte Carlo simulation; breakup process.

Se presenta un algoritmo de Monte Carlo para simular la evolución del espectro de gotas por coalescencia y rompimiento inducido por colisiones. El algoritmo estocástico de Gillespie [1] para las reacciones químicas en la formulación propuesta por Laurenzi y Diamond [2] fue utilizado para simular la cinética de la población de gotas. El rompimiento inducido por colisiones es modelado en el formalismo de Gillespie [1] como una nueva "reacción química". Los resultados fueron comparados con la solución analítica para la ecuación de rompimiento encontrada por Feingold *et al.* [3] para una distribución exponencial de las gotas satélites, y kernels de colección y rompimiento constantes. Se encontró una buena correspondencia entre la solución analítica y el algoritmo estocástico para este caso.

Descriptores: Microfísica de nubes; simulación de Monte Carlo; proceso de rompimiento.

PACS: 92.60.N; 92.60.Nv

1. Introduction

The kinetic collection equation (KCE) and the stochastic collision-breakup (SBE) equations describe the temporal change of the mean number of particles of mass x. These equations give a deterministic description of the kinetics of the drop population averaged over some volume of fluid. For the collection process the KCE has the form;:

$$\frac{\partial f(x,t)}{\partial t} = \frac{1}{2}\int_0^x f(x_1,t)f(x-x_1,t)K(x,x-x_1)dx - f(x,t)\int_0^\infty f(x_1,t)K(x,x_1)dx_1 \quad (1)$$

This equation, for a given initial spectrum $f\,(x,0)$ may be solved for $f\,(x,t)$ for all $t>0$. In (1), the collection kernel $K\,(x,x_1)$ contains the probability of collision-coalescence of two drops of masses x, x_1.

The KCE gives the time rate of change of the average number of x droplets as the difference of two terms: the first term describes the average rate of production of x droplets due to coalescence between pairs of drops whose masses sum x, and the second term describes the average rate of depletion of x droplets due to their coalescences with other droplets. Nevertheless, as was pointed out by Gillespie [4], the KCE is only an approximate time-evolution equation for $f\,(x,t)$ because the number of droplets of different masses is statistically correlated, and the KCE equation contains no definite information concerning the size of the fluctuations about the average, which would be observed in independent realizations of the coalescence stochastic process.

The combined equation for the evolution of droplet spectra through stochastic collection and breakup can be written in the form [5]:

$$\frac{\partial f(x,t)}{\partial t} = C(x,t) + B(x,t) \quad (2)$$

The first term in this equation describes the evolution of an average spectrum of drops due to the collision-collision coalescence process, and is calculated according to (1). The second term represents the time evolution of a spectrum of drops due to collision-induced breakup and is calculated with Eq. (3):

$$B\,(x,t) = \frac{1}{2}\int_0^\infty f\,(x_1,t)\,dx_1 \int_0^\infty f\,(x_2,t)\,B\,(x_1,x_2)$$

$$\times P\left(x;x_1,x_2\right)dx_2 - f\left(x,t\right)\int_0^{\infty}\frac{f\left(x_2,t\right)B\left(x,x_2\right)}{x+x_2}dx_2$$

$$\times \int_0^{x+x_2} x_1 P\left(x_1;x,x_2\right)dx_1 \qquad (3)$$

In Eq. (2) and (3) $f\left(x,t\right)$ is the drop size distribution with respect to mass x at time t. The collection and breakup kernels, $C\left(x,y\right)$ and $B\left(x,y\right)$ are calculated according to:

$$C\left(x,y\right) = K\left(x,y\right)E\left(x,y\right) \qquad (4)$$

$$B\left(x,y\right) = K\left(x,y\right)\left[1-E\left(x,y\right)\right] \qquad (5)$$

where $K\left(x,y\right)$ is the collision kernel for a drop of mass x and one of mass $y, E(x,y)$ is the coalescence efficiency for x and y. Function $P(x;x_1,x_2)$ characterizes the distribution of fragments resulting from a collision between drops of mass x_1 and x_2.

As in the collision-coalescence case, the SBE describes the evolution of an average drop spectrum. But actually, when raindrops collide, a distribution of fragments is produced. While solving the SBE these distributions are commonly parameterized using the quasi-stochastic assumption. The results of collisions are parameterized using a deterministic formula based on the average of a series of collisions. The use of this approximation is appropriate when there are sufficient raindrop interactions to justify the use of such an average.

A more realistic approach will be based on the generation of independent realizations of the collision-induced breakup process. By doing this, after a collision, a distribution of fragments is generated randomly. Within this framework, the distribution of fragments can be interpreted as a probability density function, and the average number concentration is obtained after the averaging process for a sufficiently large number of realizations of the stochastic process.

In our report, the stochastic algorithm of Gillespie [1] for chemical reactions was adopted instead of the algorithm previously elaborated for droplet populations [4]. This algorithm was reformulated to simulate the kinetic behavior of aggregating systems by Laurenzi *et al.* [6,7].

2. The Monte Carlo algorithm

In this study the stochastic algorithm developed in Ref. 1 for chemical reactions was used. This algorithm was reformulated to simulate the kinetic behavior of aggregating systems by Laurenzi *et al.* [6,7], by defining a species as a type of aggregate with a specific size and composition. In our case, species represent droplets of different sizes. A detailed description of the stochastic algorithm for multicomponent aggregation of particles can be found in Ref. 1 and 7. We summarize these results very briefly.

Within this framework, there is a unique index μ for each pair of droplets i, j that may collide. For a system with N species (S_1,S_2,...,S_N),

$$\mu \in \frac{N\left(N+1\right)}{2}.$$

The set $\{\mu\}$ defines the total collision space, and is equal to the total number of possible interactions. With this set the collision probability density function $P\left(\tau,\mu\right)$ can be determined. This quantity is defined by:

> $P(\tau,\mu)d\tau \equiv$ Probability that at time ***t*** the next collision in volume V will occur in the infinitesimal interval $(t+\tau, t+\tau+d\tau)$ and will be a μ collision.

Gillespie [1] derives this probability density function for a system of N species as

$$P\left(\tau,\mu\right)d\tau = a_\mu \exp\left(-\sum_{j=1}^{\frac{N(N+1)}{2}} a_j\tau\right) \qquad (6)$$

The functions $a_\mu$are calculated according to

$$a(i,j) = V^{-1}K(i,j)n_i n_j dt \equiv Pr\left\{\begin{array}{l}\text{Probability that two unlike particles } i \text{ and } j \text{ with populations (number of particles) } n_i \text{ and } n_j \text{ will collide within the imminent time interval}\end{array}\right\} \qquad (7)$$

$$a(i,i) = V^{-1}K(i,i)\frac{n_i\left(n_i-1\right)}{2}dt \equiv Pr\left\{\begin{array}{l}\text{Probability that two particles of the same species } i \text{ with population (number of particles) } n_i \text{ will collide within the imminent time interval}\end{array}\right\} \qquad (8)$$

The collision probability density function is the basis of the Monte Carlo algorithm. For calculating the evolution of the system, two random numbers τ and μ must be generated. Equation (4) leads directly to the answers of following questions. First, what is the probability distribution for the next time for a collision? Summing $P\left(\tau,\mu\right)d\tau$ over all μ (all possible collisions) results in

$$\begin{aligned} P_1\left(\tau\right)d\tau &= \sum_{\mu=1}^{\frac{N(N+1)}{2}} P\left(\tau,\mu\right) \\ &= \sum_{\mu=1}^{\frac{N(N+1)}{2}} a_\mu \exp\left(-\sum_{\nu=1}^{\frac{N(N+1)}{2}} a_\nu\tau\right) \\ &= \alpha\exp\left(-\alpha\tau\right)d\tau \end{aligned} \qquad (9)$$

with

$$\alpha = \sum_{\nu=1}^{\frac{N(N+1)}{2}} a_\nu$$

The probability function for reactions can be obtained in a similar way, by integrating the probability density function (pdf) $P(\tau,\mu)\,d\tau$ over all τ from 0 to ∞ results in

$$P_2(\mu) = \frac{a_\mu}{\alpha}. \tag{10}$$

Equation (6) shows that the probability of a collision with respect to time follows an exponential distribution. In order to obtain a random pair (τ, μ), according to the probability density function $P(\tau,\mu)$ we first generate a random number r_1 distributed uniformly over the interval [0,1]. Then the inversion method for obtaining random numbers is applied. In the inversion method this random number is taken as the probability of a collision in the time period τ according to $P_1(\tau)$. This probability is obtained by integrating $P_1(\tau)$ from 0 to τ:

$$r_1 = \int_0^\tau P_1(z)\,dx = \int_0^\tau \alpha\exp(-\alpha z)\,dz$$
$$= 1-\exp(-\alpha\tau) \tag{11}$$

Considering that 1-$r_1 = r_1^*$ is also a uniformly distributed random number in the interval [0,1], the time τ can be calculated from (9) in the form:

$$\tau = \frac{1}{\alpha}\ln\left(\frac{1}{r_1^*}\right) \tag{12}$$

The collision number μ is calculated similarly. A random number r_2 uniformly distributed over the interval [0,1] is generated. Then the pdf $P_2(\nu)$ (8) must be integrated over ν until the sum of the μ probability exceeds the random number r_2. The inequality for obtaining the collision index μ is of the form [1]:

$$\sum_{\nu=1}^{\mu-1} a_\nu < r_2\alpha \le \sum_{\nu=1}^{\mu} a_\nu \tag{13}$$

The former results lead to Gillespie's direct algorithm:

a) Initialize (set initial numbers of species, set t=0, set stopping criteria).

b) Calculate the function a_μ for all μ. Choose τ according to the exponential distribution

$$P_1(\tau) = \alpha\exp(-\alpha\tau)\,d\tau$$

c) Calculate μ according to the distribution

$$P_2(\mu) = a_\mu/\alpha.$$

d) Change the numbers of species to reflect the execution of a collision.

e) If stopping criteria are not met, return to step 2.

3. Treatment of the collision-induced breakup and simulation results

3.1. Stochastic treatment of the collision-induced breakup

In expressions (7) and (8), $K(i,j)$ is the collision kernel and V is the cloud volume. It is assumed that the two events, denoted by C (collision-coalescence) and B (collision-breakup), are mutually exclusive, *i.e.*, that whatever does not coalesce results in breakup. Cases of bounce are not taken into account. Then the probability of "C" or "B" is just the sum of the individual probabilities:

$$P(C\cup B) = P(C) + P(B). \tag{14}$$

The probability that two unlike particles i and j will collide within the imminent time interval can be decomposed in the sum [2]:

$$a(i,j) = V^{-1}C(i,j)n_i n_j dt + V^{-1}B(i,j)\,n_i n_j \tag{15}$$

where the first term is the probability of collection and the second the breakup probability. The collection and breakup kernels are calculated according to (4) and (5). In general, the coalescence efficiency $E(i,j)$ is based on the sizes of the colliding drops and the collisional kinetic energy [8]. The choice between coalescence and breakup can be defined from expression (13), by noting that [2]:

$$a_\mu = \alpha_\mu + \beta_\mu$$

with

$$\alpha_\mu = V^{-1}C(i,j)n_i n_j dt$$

and

$$\beta_\mu = V^{-1}B(i,j)n_i n_j dt$$

Then if β_μ causes $r_2\alpha$ to be exceeded in Eq. (13) then the event to come will be breakup; otherwise, the event will be coalescence. If the event to come is a breakup, it is calculated by randomly generating the distribution of fragments with the aid of the function $P(m,x,x_1)$.

Within the stochastic framework, the fragment distribution $P(m,x,x_1)$ can be interpreted as a probability density function. In our approach, after a collision of droplets with masses x and x_1, the satellite drops are generated as random numbers. The random generation process stops when the total mass of satellite drops exceeds the total mass of the colliding droplets. The final distribution is updated by relocating the fragments in the bins according to their mass.

Feingold *et al.* [3] found an analytical solution to the SBE equation with a constant breakup kernel and by choosing a fragment distribution of the form:

$$P(m;x,x_1) = \gamma^2(x+x_1)\exp(-\gamma m) \tag{16}$$

with $\gamma = n(N_0/M_0)$, where N_0 and M_0 are the drop number concentration and liquid water content of the initial distribution, and n is a positive integer that characterizes the fragment distribution. For the drop number concentration they found the time evolution:

$$N(t) = \frac{N_0 e^{\alpha t}}{\left(1 + \frac{N_0}{\gamma M_0}(e^{\alpha t} - 1)\right)} \tag{17}$$

where $\alpha = B\gamma M_0$, and B is the constant breakup kernel.

In this case, the fragment distribution can be interpreted as an exponential probability density function. After a collision of droplets with masses x and x_1, the satellite drops are generated as exponentially distributed random numbers.

3.2. Simulation results

3.2.1. Coalescence efficiency ***E(x,y)=1.***

The results of Monte Carlo simulations with a constant collection kernel $K(x, y)$, and consequently, constant collection and breakup kernels (expressions 4 and 5) are presented in this section by varying the coalescence efficiency $E(x, y)$. For $E(x, y)$ =1, every collision results in coalescence; for $E(x, y)$ =0, every collision between cloud droplets leads to a breakup with fragments that follow the exponential distribution (16). When $0 < E(x, y) < 1$, whatever does not coalesce results in breakup. As was remarked in Sec. 3.1, cases of bounce are not taken in account. Constant collision kernel and collection efficiencies are not realistic, but will make possible a comparison with the analytical solutions of the SCE and SBE Eqs. (1) and (3).

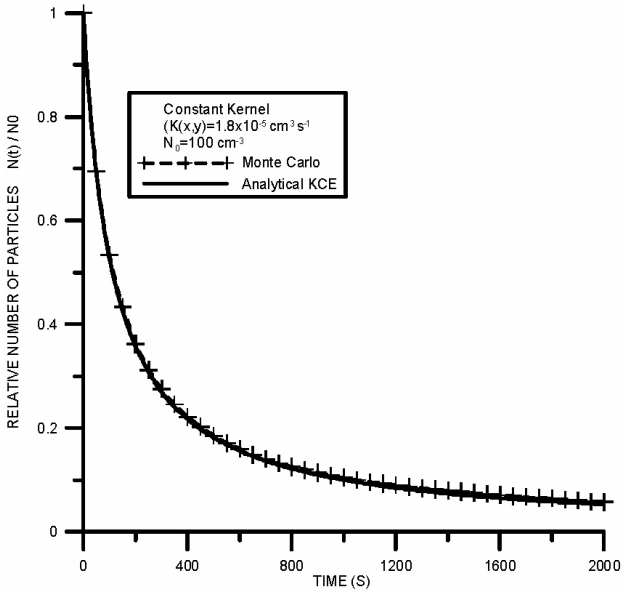

FIGURE 1. The normalized number of particles averaged over 1000 simulation runs and normalized to the initial number of particles (N_0=100) versus time is shown by the dashed line with crosses. The results from the analytical solution to the kinetic collection equation are shown by the continuous line.

TABLE I. Average rate of collision-coalescence events for a coalescence efficiency $E(x, y)$=0.8.

Average rate	Number of realizations
0.840	10
0.784	100
0.798	1000

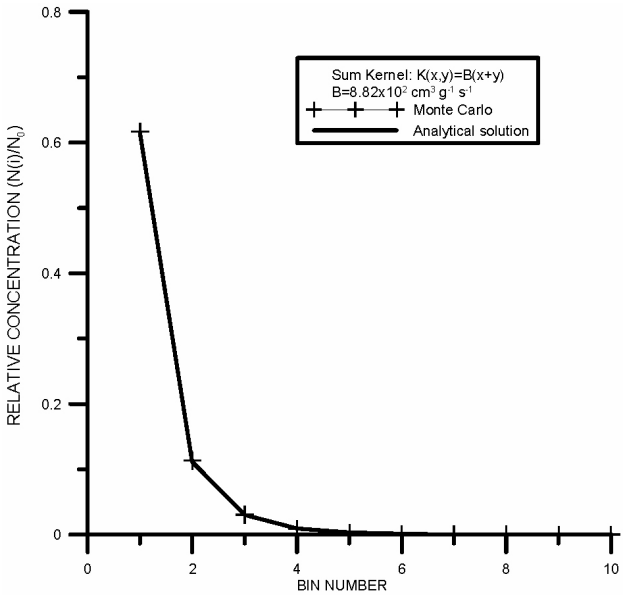

FIGURE 2. The number of particles, averaged over 1000 simulations and normalized to initial number of particles (crosses), versus the analytical solution to the kinetic collection equation (KCE) at t=700 sec. as a function of size. The initial number of particles is N_0=100.

For pure coalescence ($E(x, y)$=1), the results from the Monte Carlo algorithm are the averages over 1000 realizations of the stochastic process. In this case, every collision results in coalescence [1,7]. For monodisperse initial conditions, we consider a cloud of 1 cm^3 volume, initially containing N_0 droplets of 10 μm. These droplets were placed in bin 1 of the size distribution. We have tested our code by comparing the true stochastic averages from 1000 realizations of the stochastic algorithm and droplet concentration from the analytical solution to the KCE for the constant collection kernel (*K(x,y)*=*A*=1.8$\times 10^{-5}$cm^{3}s^{-1}) with monodisperse initial conditions derived by Scott [9] (see Fig. 1):

$$N(t) = \frac{2N_0}{T+2} \quad \text{with} \quad T = AN_0 t. \tag{18}$$

In this simulation, the initial droplet concentration was set equal to N_0=100 cm^{-3}. As can be observed, Monte Carlo simulations yielded the same results as the analytical solutions to the KCE.

For $E(x, y)$=1, the numerical code has also been tested against the analytical solution to the KCE obtained by

Golovin [10] for the sum kernel ($K(x,y)$=$B(x+y)$, B=8.82$\times$10^2 cm^{3}g^{-1} sec^{-1}) with monodisperse initial conditions:

$$N(i,t) = N_0(1-\phi)\frac{(i\phi)^{i-1}}{\Gamma(i+1)}\exp(-i\phi) \tag{19a}$$

with

$$\phi = 1-\exp(-BN_0v_0t). \tag{19b}$$

In Eq. (19 a,b), Γ is the gamma function, N_0 the initial droplet concentration, B is the constant for the Golovin kernel, v_0 is the mass of the monomer droplet (4.188$\times$10^{-9} g) and t is the time in seconds. The comparison was made for the same monodisperse initial distribution (100 droplets of 10 μm). Figure 2 depicts the size distribution for t=700 s. Again, an excellent agreement was founded between the simulated and analytical size distributions (see Fig. 2).

3.2.2. *Coalescence efficiency* $0 \leq E(x,y) < 1$

The proportion of droplets that will coalesce or break up depends on the coalescence efficiency $E(x,y)$. For example, if $E(x,y)$ =0.8, this means that on average 80% of the total number of collisions lead to coalescence, and 20% result in breakup. Table I displays the results of the average rate of coalescence-breakup events by varying the number of realizations of the stochastic process.

This average rate can be calculated as the ratio of the number of collision-coalescence events and the total number of collisions. As can be observed, the average tends to the value of 0.8 for the coalescence rate as the number of realizations is increased.

In our framework, after a collision-breakup event, the satellite drops will follow an exponential distribution. The masses of the fragments are calculated as a sequence of random numbers distributed according to the probability density function $P(m;x,x_1)$. In the present case, $P(m;x,x_1)$ has the exponential form (16). Then, the inversion method is again applied in order to obtain the masses of the satellite droplets (see the analogue expressions (11) and (12) used to obtain the exponentially distributed times to the next collision). The exponential distribution is a continuous distribution having the general form:

$$f(x) = \frac{1}{\theta}\exp\left(-\frac{x}{\theta}\right) \quad \text{with} \quad x>0 \tag{20}$$

with mean $E(X)=\theta$ and variance $V(X)=\theta^2$. Then, by comparing (16) and (20) it can be concluded that the mean mass of the satellite droplets is calculated as

$$\frac{1}{\gamma} = \frac{M_0}{nN_0}. \tag{21}$$

The average mass of the initial distribution is M_0/N_0, because n=1 in expression (21) represents the case when the average mass of the satellite droplets equals the average mass of the initial distribution. In that case there is no evolution with respect to $N(t)$ [3]. By integrating (16) with respect to m we obtain:

$$\int_0^{x+y}\gamma^2(x+y)e^{-\gamma m}dm = \frac{(x+y)}{(M_0/nN_0)} - \varepsilon(\gamma,x,y) \tag{22}$$

In (22), $x+y$ is the mass of the colliding drops and $\varepsilon(\gamma,x,y)$ is a few orders of magnitude less than unity if the number of fragments from a collision is $\geq$ 10 [3]. The numerator in the first term in the rhs of (22) is the total mass of the colliding droplets, and the denominator M_0/nN_0 is the

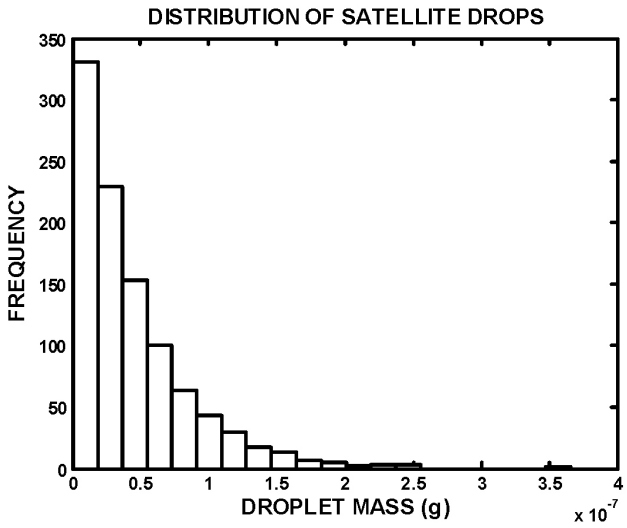

FIGURE 3. Exponentially distributed satellite droplets obtained after a random sampling with the fragment distribution. The mean mass of the distribution was set equal to 4.188$\times$10^{-8} g.

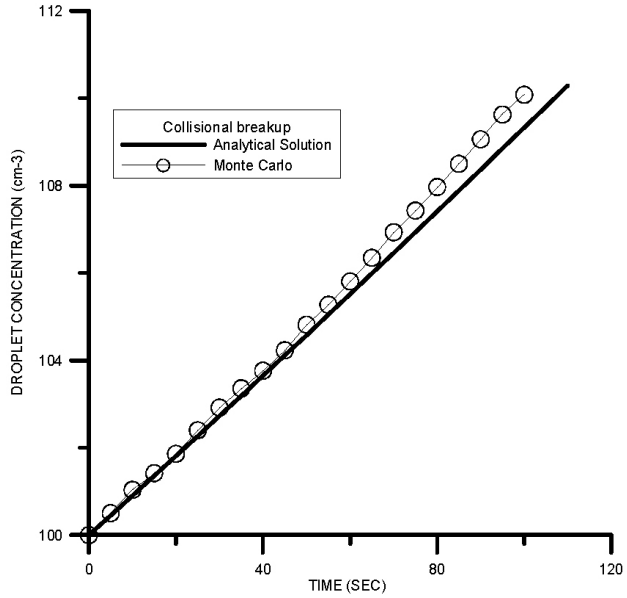

FIGURE 4. The results from the analytical solution (17) are shown by the solid line, versus the Monte Carlo results averaged over 1000 simulation runs.

average mass of the satellite droplets, so that $(x + y)/(M_0/nN_0)$ is the average number of fragments after a collision. The random sampling stops when the total mass of satellite drops exceeds the total mass of the colliding droplets. The final distribution is updated by relocating the fragments in the bins according to their mass. Figure 3 displays the exponentially distributed satellite droplets obtained after a random sampling with (16).

In order to check the algorithm for the collisional breakup case, the results from the Monte Carlo are compared with the analytical solution (17). The Monte Carlo simulation was performed with a constant breakup kernel with the exponential distribution (16) for the formed satellite drops. As the purpose of the simulation is to check the viability of the algorithm, the coalescence efficiency was set equal to 0. The initial concentration was 100 cm^{-3} (50-36.8 μm droplets and 50-37 μm droplets). The results obtained with the Monte Carlo algorithm and the analytical solution are shown in Fig. 4. As can be observed, a good correspondence between the deterministic and the stochastic process is obtained for this case.

4. Conclusions

The chemical reactions stochastic algorithm developed by Gillespie [1] was implemented in order to calculate the drop growth by collision-coalescence and collision induced breakup. Within this framework, the collision-induced breakup is introduced by considering the collision induced breakup probability as a new reaction channel. The results obtained with the Monte Carlo algorithm were compared with the analytical solutions derived by Scott [9] for the collision-coalescence process for a constant kernel. For collision-induced breakup, the Monte Carlo framework was compared with the analytical solutions derived by Feingold *et. al.* [3] for an exponential distribution of satellite drops. A very good correspondence between the Monte Carlo and analytical solutions was found for the two cases.

A further study is needed in order to include in the framework the parameterization for fragment size distribution of satellite drops developed by Low and List [8]. The advantage to the Monte Carlo approach over the analytical solution is that it does not require a specific parameterization of the fragmentation function. Given that drop breakup can be more complex that can be easily parameterized, the Monte Carlo approach will allow a more detailed study of how precipitation evolves and the sensitivity to the fragmentation function used.

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