P2.43 MONTE CARLO SIMULATIONS OF DROP GROWTH BY COALESCENCE AND COLLISION-INDUCED BREAKUP

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1. INTRODUCTION

The stochastic collection (SCE) and the stochastic collection/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 suspensión averaged over some volume of fluid. For collision/coalescence the population balance equation (PBE) is the well known SCE:

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

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

The SCE 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 (1975), the SCE is only an approximate time-evolution equation for $f(x,t)$ because the numbers of droplets of different masses are statistically correlated, and the SCE 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:

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

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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 the equation:

$$B(x,t) = \frac{1}{2}\int_0^\infty f(x,t)dx \int_0^\infty f(x_1,t) B(x,x_1) \times P(m;x,x_1) dx_1 - f(m,t)\int_0^\infty \frac{f(x_1,t) B(m,x_1)}{m+x_1} \times \int_0^{m+x_1} xP(x;m,x_1) dx \quad (3)$$

The breakup equation is written in the form proposed by Gillespie and List (1976). The collection and breakup kernels, $C(x,x_1)$ and $B(x,x_1)$ are calculate according to:

$$C(x,x_1) = K(x,x_1) E(x,x_1) \quad (4)$$

$$B(x,x_1) = K(x,x_1)\left[1 - E(x,x_1)\right] \quad (5)$$

here $K(x,x_1)$ is the collision kernel for a drop of mass *x* and one of mass x_1, $E(x,x_1)$ is the coalescence efficiency for *x* and x_1. Function $P(m;x)$ characterizes the distribution of fragments.

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 averaged 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 is a probability density functions, 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 (1976) for chemical reactions was adopted

instead of the algorithm previously elaborated for droplet populations (Gillespie, 1975). This algorithm was reformulated to simulate the kinetic behaviour of aggregating systems by Laurenzi and Diamond (1999).

2. THE MONTE CARLO ALGORITHM

In our report, the stochastic algorithm of Gillespie (1976) for chemical reactions was adopted instead of the algorithm previously elaborated for droplet populations (Gillespie, 1975). This algorithm was reformulated to simulate the kinetic behaviour of aggregating systems by Laurenzi and Diamond (1999). In Laurenzi and Diamond (1999) a species are defined as a type of aggregate with a specific size and composition, in our specific case, species are defined as droplets of specific size.

Within this framework, there is a unique index μ for each pair of droplets *i, j* that may react (collide). For a system with *N* species $(S_1,\ S_2,\ \ldots\ ,S_N)$ $\mu \in \frac{N(N+1)}{2}$. The set $\{\mu\}$ defines the total "collision" space, and is equal to the total number of possible interactions (collisions). With this set the reaction probability density function $P(\tau,\mu)$ can be determined.This quantity is defined by

$P(\tau,\mu)d\tau \equiv$ Probability that at time the next reaction (collision) in volume V will occur in the infinitesimal interval $(t+\tau,\ t+\tau+d\tau)$ and will be a μ reaction. In Gillespie (1976) this probability density function has been derived for a system of *N* species as

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

Here $\mu \in \frac{N(N+1)}{2}$. The functions a_μ are calculated according to

$a(i,j) = V^{-1}K(i,j)X_iX_j dt$ =Pr{ Probability that two unlike particles *i* and *j* with populations (number of particles) X_i and X_j will collide within the inminent time interval} (7)

$$a(i,i) = V^{-1}K(i,i)\frac{X_i(X_i-1)}{2}dt$$ Pr{Probability that two particles of the same species *i* with population (number of particles) X_i collide within the inminent time interval} (8)

The reaction 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 (11) leads directly to the answers of the aforementioned questions. First, what is the probability distribution for times?. Summing $P(\tau,\mu)d\tau$ over all μ (all possible collisions, (reactions)) results in

$$P_1(\tau)d\tau = \sum_{\mu=1}^{\frac{N(N+1)}{2}} a_\mu \exp\left(-\sum_{\nu=1}^{\frac{N(N+1)}{2}} a_\nu\tau\right)$$

$$= \alpha\exp(-\alpha\tau)d\tau \qquad (12)$$

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 pdf $P(\tau,\mu)d\tau$ over all τ from 0 to ∞ results in

$$P_2(\mu) = \frac{a_\mu}{\alpha} \qquad (13)$$

Equation (13) gives the probability of a particular reaction μ given an interval (τ, $\tau+d\tau$). Equation (12) shows that the probability of a reaction (collision) in time follows an exponential distribution, a characteristic of a process in which events occurs randomly in time.

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 in the interval [0,1], then, the inversion method to obtain random numbers is applied. In the inversion method this random number is taken as the probability of a reaction in the time period τ according to $P_1(\tau)$. This probability is obtained by integrating $P_1(\tau)$ from 0 to τ:

$$r_1 = \int_0^\tau P_1(x)dx = \int_0^\tau \alpha\exp(-\alpha x)dx = 1-\exp(-\alpha\tau) \qquad (14)$$

Considering that $1-r_1=r_1^*$ is also a uniformly distributed random number in the interval [0,1], then the time τ can be calculated from (14) in the form:

$$\tau = \frac{1}{\alpha}\ln\left(\frac{1}{r_1^*}\right) \qquad (15)$$

The reaction number μ is calculated similarly. A random number r_2 uniformly distributed in the interval [0,1] is generated. Then the pdf $P_2(\nu)$ (13) must be integrated over v until the addition of the μ probability exceeds the random number r_2. The inequality to obtain the reaction index μ has the form (Gillespie, 1976)

$$\sum_{\nu=1}^{\mu-1} a_\nu < r_2\alpha \le \sum_{\nu=1}^{\mu} a_\nu \qquad (16)$$

The former results lead to the Gillespie's direct algorithm:

1) Initialize (set initial numbers of species, set t=0, set stopping criteria).
2) Calculate the function a_μ for all μ.

3) Choose τ according to the exponential distribution $P_1(\tau) = \alpha \exp(-\alpha\tau) d\tau$
4) Calculate μ according to the distribution $P_2(\mu) = \frac{a_\mu}{\alpha}$.
5) Change the numbers of species to reflect the execution of a reaction.
6) If stopping criteria are not met, go to step 2.

2.1 TREATMENT OF THE COLLISION-INDUCED BREAKUP

In the 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 (breakup) are mutually exclusive, i.e., that whatever does not coalesce results in breakup. Then the probability of "C" or "B" is just the sum of the individual probabilities.

$$P(CUB) = P(C) + P(B) \quad (9)$$

Then, the probability that two unlike particles *i* and *j* will collide within the inminent time interval can be decomposed in the sum:

$$a(i, j) = V^{-1} C(i, j) X_i X_j dt + V^{-1} B(i, j) X_i X_j \quad (10)$$

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 (Low and List, 1982a). The choice between coalescence and breakup can be defined from expression (16), by noting that

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

with

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

and

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

Then if β_μ causes $r_2\alpha$ to be exceeded in equation (16) then the event to come will be breakup, otherwise, the event will be a coalescence.

Is the event to come is a collisional breakup event, then 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. 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 obtained by relocating the fragments in the bins according to its mass.

Feingold et al. (1988) found an analytical solution of the SBE equation with a constant breakup kernel and by choosing a fragment distribution of the form:

$$P(m; x, x_1) = \gamma^2 (x + x_1) \exp(-\gamma m) \quad (11)$$

with $\gamma = n \frac{N_0}{M_0}$ where N_0 and M_0 are the drop number concentration and liquid water content of the initial distribution. For the drop number concentration they found the time evolution:

$$N(t) = \frac{N_0 e^{\alpha t}}{\left(1 + \frac{N_0}{\gamma M_0}(e^{\alpha t} - 1)\right)} \quad (12)$$

where $\alpha = B\gamma M_0$

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. The performance of the algorithm can be checked by a direct comparison with the analytical solution (12).

3. SIMULATION RESULTS

For pure coalescence, the results from the Monte Carlo algorithm are the averages over 1000 realizations of the stochastic process. 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. Fig.1 shows a comparison between the Monte Carlo algorithm and an analytical solutions of the SCE for the contant kernel. The monodisperse initial distribution was set equal to N_0=100 cm^{-3}. As can be observed simulations, yielded the same results as the analytical solutions of the SCE.

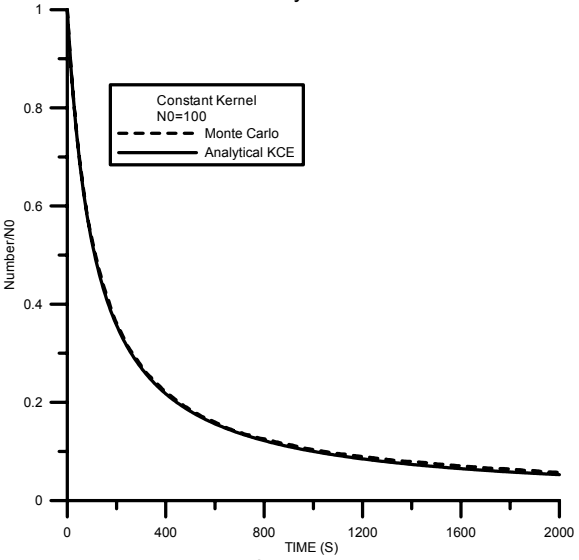

Fig. 1. The number of particles averaged over 1000 simulation runs and normalized to the initial number of particles (N_0=100), versus time is shown by the dashed line. The results from the analytical solution are shown by the solid line.

In order to check the algorithm for the collisional breakup case, the results from the Monte Carlo are compared with the analytical solution (12).

The Monte Carlo simulation was performed with a constant breakup kernel with the exponential distribution (11) for the formed satellite drops. 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 Figure 2. As can be observed, a good correspondence between the deterministic and the stochastic process is obtained. A further increase in the number of realizations for this case is needed in order to reduce the fluctuations associated with the Monte Carlo algorithm.

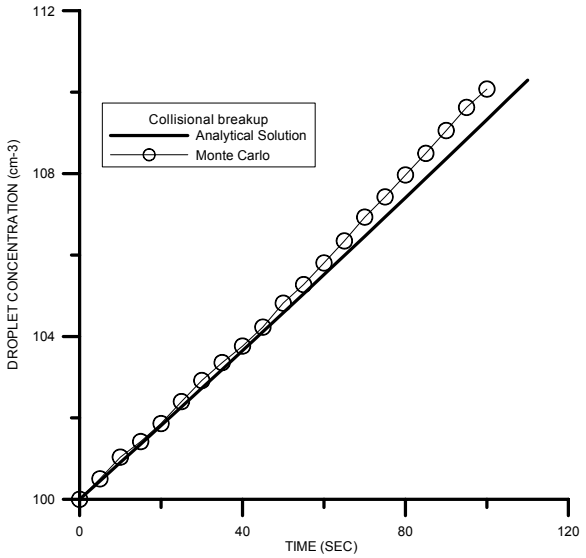

Fig. 2. The results from the analytical solution (12) are shown by the solid line, versus the Monte Carlo results averaged over 1000 simulation runs.

4. CONCLUSIONS

The chemical reactions stochastic algorithm developed by Gillespie (1976) 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 (1968) for the collision-coalescence process for a constant kernel. A very good correspondence between the Monte Carlo and the deterministic solutions were founded.

For collision-induced breakup, the Monte Carlo framework was compared with the analytical solutions derived by Feingold et. al (1988). Although a good correspondence was obtained, a further increase in the number of realizations is needed in order to reduce the random fluctuations inherent to the stochastic process.

5. REFERENCES

Feingold, G., S. Tzivion and Z. Levin, 1988: The evolution of raindrop spectra. Part I. Stochastic collection and breakup. J. Atmos. Sci., 45, 3387-3399.

Gillespie, D.T. 1975. An exact method for numerically simulating the stochastic coalescence process in a cloud. *J . Atmos. Sci*. 32: 1977-1989.

Gillespie, D.T. 1976. A general method for numerically simulating the stochastic time evolution of coupled chemical reactions. *J. Comput. Phys*. 22: 403-434.

Laurenzi, I.J. and Scott L. Diamond. Monte Carlo Simulation of the Heterotypic Aggregation Kinetics of Platelets and Neutrophils. *Biophys.* J. 77:1733-1746. 1999

Laurenzi, I.J. and S. L. Diamond. Kinetics of random aggregation-fragmentation processes with multiple components. Phys. Rev. E. 67:051103. 2003.

List, R. and J.R. Gillespie, 1976: Evolution of Raindrop Spectra with Collision-Induced Breakup. *J. Atmos. Sci.,* 33, 2007–2013.

Low, T.B. and R. List, 1982: Collision, Coalescence and Breakup of Raindrops. Part II: Parameterization of Fragment Size Distributions. J. Atmos. Sci., Vol. 39, No. 7, pp. 1607–1619.

Scott, W.T., 1968: Analytic studies of cloud droplet coalescence. J. Atmos. Sci. 25, 54-65.