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Atmospheric Chemistry and Physics Discussions

Monte Carlo simulations of two-component drop growth by stochastic coalescence

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Abstract

The evolution of two-dimensional drop distributions is simulated in this study using a Monte Carlo method. The stochastic algorithm of Gillespie (1976) for chemical reactions in the formulation proposed by Laurenzi et al. (2002) was used to simulate the kinetic behavior of the drop population. Within this framework species are defined as droplets of specific size and aerosol composition. The performance of the algorithm was checked by comparing the numerical with the analytical solutions found by Lushnikov (1975). Very good agreement was observed between the Monte Carlo simulations and the analytical solution.

Simulation results are presented for bi-variate constant and hydrodynamic kernels. The algorithm can be easily extended to incorporate various properties of clouds such as including several crystal habits, different types of soluble CCN, particle charging and drop breakup.

1 Introduction

The understanding of aerosol-cloud interactions contains large uncertainties that must be reduced to accurately estimate the impact of aerosols on weather and climate. One of the most problematic aspects of aerosol-cloud interactions is the collision-coalescence process that is a mechanism that modifies the aerosol distribution, i.e. the aerosol particles that are the nuclei for individual droplets are combined during the coalescence process in the same way as the mass of the individual water droplets are merged. After the evaporation of the drop formed by coalescence, the aerosol particle that remains will have the mass of the original two nuclei.

The aerosol distribution becomes important as the cloud drops evaporate and the solutes are recycled into aerosols that can serve as CCN: the larger the mass of a hygroscopic aerosol, the lower the supersaturation needed to form a cloud droplet. In the marine environment, the aerosol recycling process is believed to be the major mech-

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anism responsible for the bimodal shape of the aerosol size distributions (Flossmann, 1994; Feingold et al., 1996). The heterogeneous chemical reactions, which add non-volatile solute to each cloud droplet, strongly depend on the salt content and pH of the droplet (Alfonso and Raga, 2004). Since aerosols also have a significant influence on cloud microphysics and cloud radiative properties, it is necessary to simulate aerosol processes realistically and with adequate accuracy.

In general, cloud models with detailed microphysics describe the aerosol and cloud droplets with two separate one-dimensional size distributions. With this approach only the average aerosol mass contained in cloud droplets of a particular size is predicted by the model and is not possible to keep track of the spectral aerosol mass distribution within the cloud droplets. For the deterministic case, the aerosol processing due to collision-coalescence was addressed by Liu (1998) and Bott (2000) by extending the flux method to two-dimensional distributions. Within this framework each particle is characterized both by the mass of its dry aerosol nucleus and by its water mass. Nevertheless, an extension of the exact stochastic framework developed by Gillespie (1976) for a two parameter droplet spectrum has never been reported in the cloud physics literature.

The main advantage of the stochastic approach, described in this paper, over deterministic methods is that it can be easily extended to include not only the solute mass, but other particle properties such as crystal habit, different populations of CCN, chemical composition and the breakup of droplets (Alfonso et al., 2006).

Here we apply the general multi-component algorithm described by Laurenzi et al. (2002) to the solution of the kinetic collection equation (KCE) in cloud models dealing with two-dimensional microphysics.

The discrete two-component KCE, which is an extension of the discrete one dimensional kinetic collection equation, is given as:

$$\frac{\partial N(m,n;t)}{\partial t} = \frac{1}{2}\sum_{m'=0}^{m}\sum_{n'=0}^{n} K(m-m', n-n'; m', n'; t)N(m-m', n-n'; t)N(m', n', t)$$

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$$-N(m,n;t)\sum_{m'=0}^{\infty}\sum_{n'=0}^{\infty}K(m,n;m',n')N(m',n';t) \quad (1)$$

Where $N(m,n;t)$ is the average number of species with water mass from size bin m and aerosol mass from size bin n. The water mass in size bin m equals the volume of a droplet in the smallest (monomer droplet) bin multiplied by m, the aerosol mass in size bin n equals the volume of an aerosol in the smallest bin (monomer aerosol) multiplied by n. In general, $N(m,n;t)$ is the average number of particles consisting of m monomers of the first and n of the second kind, respectively. The integral (continuous) version of this equation is more familiar:

$$\frac{\partial N(n,m;t)}{\partial t} = \frac{1}{2}\int_0^m dm_1 \int_0^n dn_1 K(m-m_1,n-n_1;m_1,n_1)N(m-m_1,n-n_1;t)N(m_1,n_1;t)$$

$$-N(m,n;t)\int_0^{\infty} dm_1 \int_0^{\infty} dn_1 K(m,n;m_1,n_1)N(m_1,n_1;t) \quad (2)$$

In Eqs. (1) and (2) $K(m,n;m_1,n_1)$ is the collection kernel, now dependent on the composition of coagulating particles. The discrete KCE Eq. (1) gives the time rate of change of the average number of species with water mass from bin m and aerosols from bin n as the difference of two terms, the first term describes the average rate of production of the (m,n) species due to coalescence between pairs of particles whose water mass volume is in size bin m, and the aerosol volume is in size bin n and the second term describes the average rate of depletion of (m,n) particles due to their coalescence with particles from other species. To solve Eqs. (1) and (2) initial conditions are needed:

$$N(m,n;0) = N_0(m,n) \quad (3)$$

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For the discrete case, we also put $N(0,0;t)=0$ for every t. The numerical solution of the KCE Eqs. (1) and (2) is difficult due to the double integral and nonlinear behavior of the equation and several numerical techniques can be found in the literature. In cloud physics modeling, Eq. (2) was numerically integrated by the flux method developed by Bott (2000) and independently by Liu (1998), both assuming that the probability for the collision of two cloud droplets depends only on the water mass of each one and not on the mass of the aerosol nuclei.

Other methods are computationally more expensive, such as the previously mentioned Monte Carlo (MC) algorithm developed by Laurenzi et al. (2002). This method has the advantage that it can be employed to determine both the expectations and fluctuations for multi-component aggregation. On the other hand, the KCE may not be valid at longer time periods, when a single drop acquires a mass much larger than the rest of the population and becomes separated from the continuous mass spectrum. In such a situation, the statistical fluctuations at the high-mass end of the spectrum must be taken into account. The Monte Carlo method is also very useful while investigating the role of coalescence in redistributing the aerosol mass in early warm rain stages when the artificial broadening of the drop distribution must be avoided.

2 The stochastic algorithm

A detailed description of the stochastic algorithm for multi-component aggregation of particles can be found in Gillespie (1976) and Laurenzi et al. (2002), and we briefly summarize it here. Consider a well-mixed and spatially homogeneous volume V in which particles belonging to N_s distinct species are present. Each species is characterized both by its water mass and by the mass of its dry aerosol nucleus, $\bar{u}_\mu = (u_m, u_n)$, such that, a droplet with composition $\bar{u}_\mu$ is a member of the μth species. After time $t=0$ the species will randomly coalesce according to:

$$A_{m,n} + B_{m',n'} = C_{m+m',n+n'} \tag{4}$$

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where $A_{m,n}$ and $B_{m',n'}$ are droplets with compositions $\bar{u}_\mu = (u_m, u_n)$ and $\bar{u}_\nu = (u_{m'}, u_{n'})$, respectively. The transition probabilities for coalescence events follow Laurenzi et al. (2002) and are given by:

$$a(i,j) = V^{-1}K(i,j)n_i n_j dt \equiv \Pr\{\text{Probability that two particlesof species } i \text{ and } j \text{ (for } i \neq j\text{) with populations (number of particles) } n_i \text{ and } n_j \text{ will collide within the inminent time interval}\} \quad (5)$$

$$a(i,i) = V^{-1}K(i,i)\frac{n_i(n_i-1)}{2}dt \equiv \Pr\{\text{Probability that two particles of the same species } i \text{ with population (number of particles) } n_i \text{ collide within the inminent time interval}\} \quad (6)$$

In Eqs. (5) and (6), $K(i,j)$ is the collection kernel, and V is the cloud volume. 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 $(S_1, S_2, \ldots, S_N)$ $\nu \in \frac{N(N+1)}{2}$. The set $\{\nu\}$ defines the total collision space, and is equal to the total number of possible interactions. The transition probabilities Eqs. (5) and (6) are then represented by one index (a_ν).

This stochastic model is solved using the algorithm introduced by Gillespie (1976) for chemical kinetics and modified by Laurenzi et al. (2002). The expected behavior of the system can be evaluated by averaging over many realizations of the stochastic process, described by the following steps:

1) At t=0, the event counter is set to zero and the initial number of species $n_1, n_2, \ldots, n_N$. is defined

2)The quantity α is calculated as:

$$\alpha = \sum_{\nu=1}^{\frac{N(N+1)}{2}} a_\nu \quad (7)$$

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Generate a random number r_1 from a uniform distribution in the interval (0,1) and considering that 1-r_1=r_1^* is also a uniformly distributed random number calculate

$$\tau = \frac{1}{\alpha} \ln\left(\frac{1}{r_1^*}\right) \tag{8}$$

3) Generate a random number r_2 from a uniform distribution in the interval (0,1). Choose a collision ("chemical reaction") with index μ from the inequality

$$\sum_{\nu=1}^{\mu-1} a_\nu < r_2\alpha \leq \sum_{\nu=1}^{\mu} a_\nu \tag{9}$$

4) Let t=t+τ

5) Change the number of species to reflect the execution of collision.

3 Model results

3.1 Comparison of the Monte Carlo algorithm with analytical solutions

In order to check the performance of the Monte Carlo algorithm, a simulation with a constant kernel was performed and compared with the analytical solution found by Lushnikov (1975). Solutions to Eqs. (1) and (2) can be obtained for an important class of collection kernels, such as when the kernel depends only on the total number of monomers (droplets and aerosols) in each colliding particle. In this case:

$$K(m, n; m_1, n_1) = K(m + n; m_1 + n_1) \tag{10}$$

Lushnikov constructed an explicit form for the composition distribution for this type of kernel, which corresponds to coagulation of initially monomeric particles. In this case $N(1, 0; 0)$=c_1 and $N(0, 1; 0)$=c_2, corresponding to the situation with initially c_1 droplets and c_2 aerosols. The composition distribution may be expressed as (Lushnikov, 1975):

$$N(m, n; t) = \binom{m+n}{n} \left(\frac{c_1}{c_0}\right)^m \left(\frac{c_2}{c_0}\right)^n N(m+n, t) \quad c_0 = c_1 + c_2 \tag{11}$$

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Where $\binom{m+n}{n}$ are the binomial coefficients, and $N(m+n,t)$ is the number of particles composed of $(m+n)$ monomers (m monomer droplets and n monomer aerosols). Lushnikov (1975) showed that $N(m+n,t)$, for the type of kernels Eq. (10) is a solution of the one dimensional kinetic collection equation:

$$\frac{\partial N(i,t)}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1} K(i-j,j)N(i-j)N(j) - N(i)\sum_{j=1}^{\infty} K(i,j)N(j) \tag{12}$$

In this case, $N(i,t)=\sum_{m+n=i}N(m,n;t)$. The initial condition for Eq. (12) is $N(i,t)=N_0\delta_{i,1}$. Analytical solutions of the continuous KCE have been obtained by Golovin (1963), Scott (1968), Drake (1972) and Drake and Wright (1972) for approximations of the hydrodynamic kernel given by the polynomials $K(i,j)=A, B(x_i+x_j)$ and $C(x_ix_j)$ where x_i and $x_j$are the masses of the droplets from bins i and j. For the constant kernel $K(i,j)=A$ and a monodisperse initial distribution with concentration c_0, the analytical size distribution of the discrete KCE has the form:

$$N(i,t) = 4c_0\frac{(T)^{i-1}}{(T+2)^{i+1}} \text{ with } T = Ac_0t \tag{13}$$

Then, the analytical solution of Eq. (1), calculated according to the expression Eq. (11) for the constant kernel $K(x_i,x_j)=A$, is compared with true stochastic averages over N_r realizations of the stochastic process (in our simulations $N_r=1000$):

$$\langle N(m,n;t)\rangle = \frac{1}{N_r}\sum_{r=1}^{N_r} N(m,n;t)^r \tag{14}$$

where $N(m,n;t)^r$ is the number of particles for species with droplet mass from bin number m and dry aerosol mass from bin n in the $r-$realization of the stochastic algorithm at time t.

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The Monte Carlo simulation was conducted for initially monomeric particles (droplets and aerosols) with concentrations c_1=30 and c_2=30 ($N(1,0;0)$=30 and $N(0,1;0)$=30). Long (1974) calculated the coefficients for the polynomials $K(x,y)=A$, $B(x+y)$ and $C(xy)$ approximating the one dimensional collection kernel when the largest of the colliding drops is smaller than 50 μm. For the constant kernel, he found a value of $A=1.20\times10^{-4}$ ($cm^3\,s^{-1}$). We used the same value for the constant discrete two-dimensional collection kernel:

$$K(m,n;m',n') = 1.2 \times 10^{-4} (\mathrm{cm}^3\,\mathrm{s}^{-1}) \quad (15)$$

In our simulations, the monomer droplet is 10 μm in radius (droplet mass 4.188×10^{-9}g) and the monomer aerosol is an ammonium sulfate aerosol, 0.1 μm in radius (aerosol mass 1.14×10^{-14}g). The aerosol-water mass grid was chosen according to Droplet_mass(i)=i × m_0 (i=1, .., N_{droplets}) and Aerosol_mass(j)=j × n_0 (j=1, .., N_{aerosols}). Here m_0 and n_0 are the masses of the monomer droplet (4.188×10^{-9}g) and the monomer aerosol (1.14×10^{-14}g) respectively.

We have defined 30 bins for the water mass grid and 30 bins for the aerosol grid. The pure monomeric species are also considered (those containing pure droplets and pure aerosols). Then, the total number of species in our numerical experimental can be calculated as:

$$N_{\mathrm{Total}} = N_{\mathrm{droplets}} \times N_{\mathrm{aerosols}} + N_{\mathrm{droplets}} + N_{\mathrm{aerosols}} \quad (16)$$

Where N_{droplets} and N_{aerosols} are the number of bins for the water mass grid and the aerosol grid respectively. The last two terms in Eq. (16) account for the monomeric species (droplets and aerosols). In our case the total number of species is 960.

The solutions obtained from the Monte Carlo calculations (averaged over 1000 realizations) for the species *N(1,1;t), N(0,1;t)* and *N(0,1;t)* are shown in Fig. 1. The analytical solution are also shown in Fig. 1 (represented by the solid curve), and indicates the good agreement between these solutions of the KCE (Eq. 1).

The two dimensional discrete size distributions for a) the analytical solution given by Eq. (11) and b) the average over 1000 realizations after 100 s are displayed in Figs. 2

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and 3. The same comparison is shown after 200 (Figs. 4 and 5) and 400 s in (Figs. 6 and 7), respectively. Note that the differences between the Monte Carlo averages and the analytical solution of the KCE are again negligible.

The one-dimensional distribution, which is a solution of the one-dimensional kinetic collection Eq. (12), can be obtained from the two-dimensional spectrum by integrating over the aerosol grid for any point in time, as:

$$N(m,t) = \sum_{n=1}^{N_{\text{aerosols}}} N(m,n;t), \quad m = 1,\ldots,N_{\text{droplets}} \tag{17}$$

In Eq. (17) N_{aerosols}, and N_{droplets} are the number of bins (grid points) in the aerosol and water grid, respectively. Two other simulations were performed with different initial conditions: $N(1,1;0)$=100 and $N(1,2;0)$=150**,** corresponding initially to 100 and 150 particles per cubic centimeter from species $(1,1)$ and $(1,2)$, respectively. From Eq. (17), a monodisperse initial condition for the one-dimensional KCE can be obtained from the two-dimensional initial condition as:

$$N(1;0) = N(1,1;0) + N(1,2;0) = 250 \tag{18}$$

For this particular case (constant kernel and monodisperse initial conditions) we can use the analytical solution Eq. (13) of the KCE in order to compare with the two-component Monte Carlo. The drop size distributions calculated from the Monte Carlo, which is obtained by integrating the particle distribution over the aerosol grid according to Eq. (17), and the analytical solution of the KCE with constant kernel (A=1.20×10^{-4} cm^3 s^{-1}) from a monodisperse initial condition N_0Eq. (1)=250 cm^{-3} are displayed in Figs. 8 and 9. Again, a good agreement between the two approaches is found.

As was remarked in detail by Laurenzi et al. (2002), the species accounting formalism outlined in Sect. 2 reduces both computer storage and simulation time. This process is handled by dynamic allocation of memory permitting calculations with thousands of droplets in the initial distribution.

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3.2 Simulations with realistic initial distributions and hydrodynamic kernel

Simulations with the two-dimensional Monte Carlo were performed with realistic initial particle distributions and with the two-dimensional hydrodynamic kernel which is relevant to cloud physics. The two-dimensional extension of the piecewise approximation found by Long (1974) was used:

$$K(i,j) = 9.44 \times 10^9 \left(x_{\text{species}}(i)^2 + x_{\text{species}}(j)^2 \right) \text{ if } R \leq 50\mu m \tag{19}$$

or by

$$K(i,j) = 5.78 \times 10^3 \left(x_{\text{species}}(i) + x_{\text{species}}(j) \right) \text{ if } R > 50\mu m \tag{20}$$

In Eqs. (19) and (20) R is the species radius and $x_{\text{species}}(i)$ is the mass of the particle from species with index i which is calculated as:

$$x_{\text{species}}(i) = x_d(i) + x_a(i) \tag{21}$$

where $x_d(i)$ and $x_a(i)$ are the droplet and the aerosol mass respectively.

Figure 10 shows the initial two-component spectrum for our simulation. The spectrum has a droplet concentration of 158 cm^{-3}. This distribution was obtained (following Liu, 1998) by assuming a gamma distribution function for the drop coordinate and an exponential distribution for the aerosol size coordinate. The Figs. 11 and 12 display the drop and aerosol distributions, averaged over 1000 realizations, for two durations (t=150 s, t=1500 s). There is a net loss for small particles and net gain for large particles. The spectrum shifts toward particles with large drop sizes and large aerosol sizes.

4 Discussion and conclusions

The multi-component MC algorithm proposed by Laurenzi et al. (2002) and based upon Gillespie's (1976) stochastic approach to chemical reactions was implemented to simulate two-component droplet growth by stochastic coalescence. Within this framework,

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all assumptions included in the stochastic collection equation are avoided. Additionally it permits calculation of statistical fluctuations for two-component droplet aggregation. On the other hand, the continuous KCE may not be valid when a single drop acquires a mass much larger than the rest of the system and becomes separated from the smooth mass spectrum. In such a situation, the statistical fluctuations at the high-mass end of the spectrum must be taken into account.

For the two-dimensional case each species is characterized both by its water mass and by the mass of its dry aerosol nucleus. Very good agreement was observed between analytical solutions of the KCE and MC simulations.

Moreover, the above described algorithm can be easily extended to the multi-component case in order to include various other properties of clouds as well as the breakup of droplets (Alfonso et al., 2006). In a more general case species can be defined as types of particles with several attributes (droplet radius, CCN composition, chemical composition, electric charge, etc.). For this case, the state of a k component system is defined by a set of drops with properties or compositions $\bar{u}_i = \left(u_{1,i}, u_{2,i}, u_{3,i}, \ldots, u_{k,i}\right)$ where $u_{k,i}$ denotes the amount of the component or the property k in species i. For example, for the ice phase, it may represent the crystal habit, or the ice crystal mass. Then, the transition probability (5) may be defined as the probability that a specific pair of particles (drops, ice crystals, aerosols) with set of properties $\bar{u}_i = \left(u_{1,i}, u_{2,i}, u_{3,i}, \ldots, u_{k,i}\right)$ and $\bar{u}_j = \left(u_{1,j}, u_{2,j}, u_{3,j}, \ldots, u_{k,j}\right)$ will aggregate in the next time interval.

The stochastic approach should make more feasible the modeling of highly complicated microphysical processes and offers a method to evaluate these processes in much greater detail than has been previously possible.

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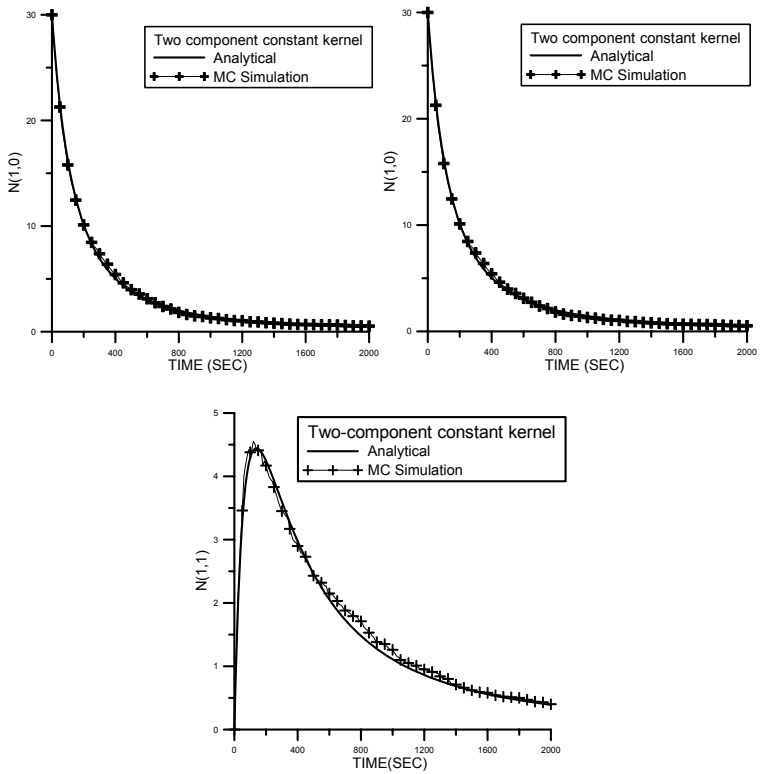

Fig. 1. Simulated time evolution of species **(a)** $N(1,0)$, **(b)** $N(0,1)$ and **(c)** $N(1,1)$ for a system modeled by the constant kernel, as a function of time. The solid lines are the analytical solutions of the two-dimensional KCE.

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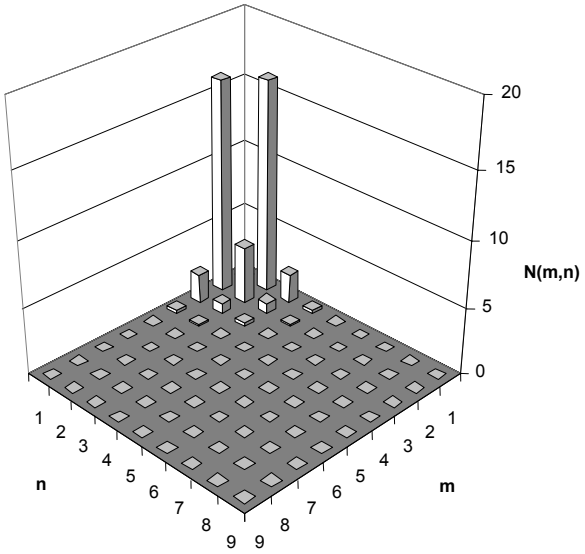

Fig. 2. Discrete two dimensional droplet distributions $N(m, n)$ resulting from the analytical solution of the two-dimensional KCE with a constant kernel at t=100 s, with monomeric initial conditions: ($N(1, 0; 0)$=30 and $N(0, 1; 0)$=30).

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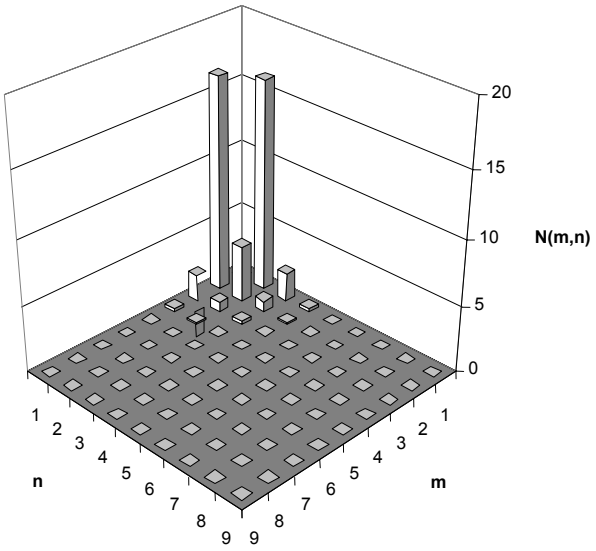

Fig. 3. Discrete two dimensional droplet distributions $N(m, n)$ resulting from the numerical solution of the two-dimensional KCE with a constant kernel at t=100 s. Monte Carlo simulations were conducted with initial conditions $N(1, 0; 0)$=30 and $N(0, 1; 0)$=30.

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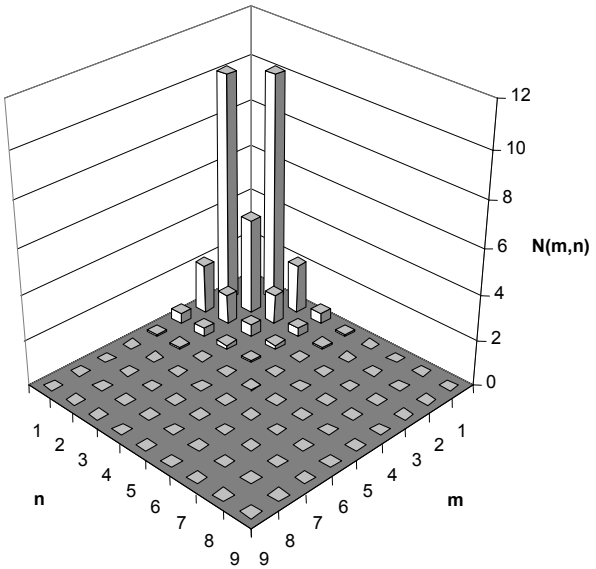

Fig. 4. Discrete two dimensional droplet distributions $N(m, n)$ resulting from the analytical solution of the two-dimensional KCE with a constant kernel at t=200 s, with monomeric initial conditions ($N(1, 0; 0)$=30 and $N(0, 1; 0)$=30).

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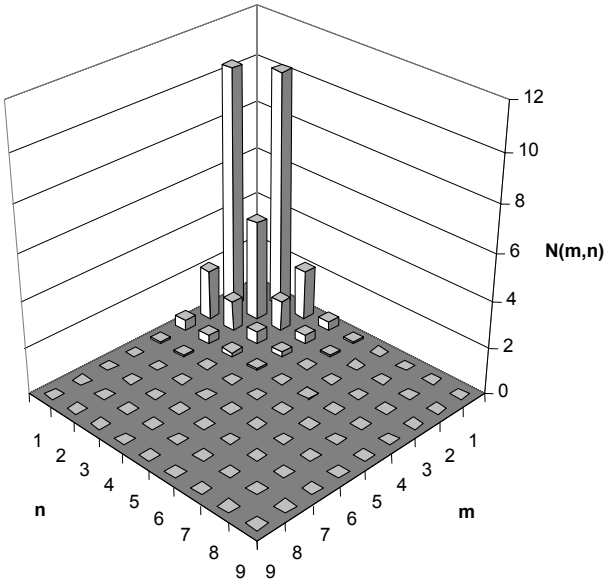

Fig. 5. Discrete two dimensional droplet distributions $N(m, n)$ resulting from the numerical solution of the two-dimensional KCE with a constant kernel at t=200 s. Monte Carlo simulations were conducted with initial conditions ($N(1, 0; 0)$=30 and $N(0, 1; 0)$=30).

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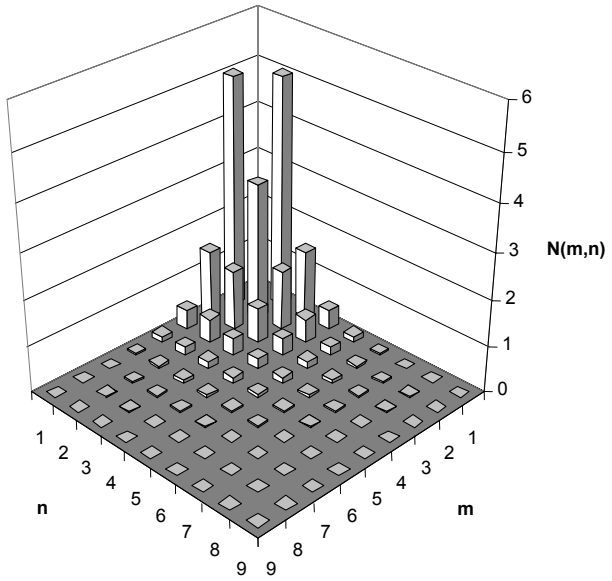

Fig. 6. Discrete two dimensional droplet distributions $N(m, n)$ resulting from the analytical solution of the two-dimensional KCE with a constant kernel at t=400 s, with monomeric initial conditions ($N(1, 0; 0)$=30 and $N(0, 1; 0)$=30).

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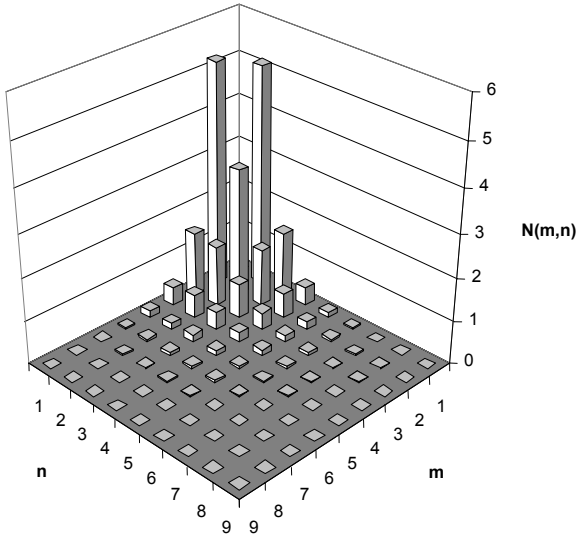

Fig. 7. Discrete two dimensional droplet distributions $N(m, n)$ resulting from the numerical solution of the two-dimensional KCE with a constant kernel at t=400 s. Monte Carlo simulations were performed with initial conditions $N(1, 0; 0)$=30 and $N(0, 1; 0)$=30.

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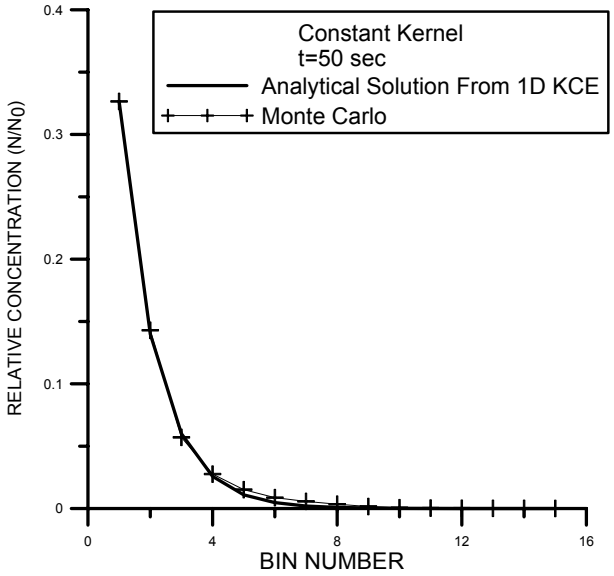

Fig. 8. The number of particles averaged over 1000 realizations and normalized to initial number of particles (N_0=250) represented by the line with crosses) and the analytical solution of the one dimensional kinetic collection equation (KCE) (represented by the dark solid line) as a function of size for t=50.

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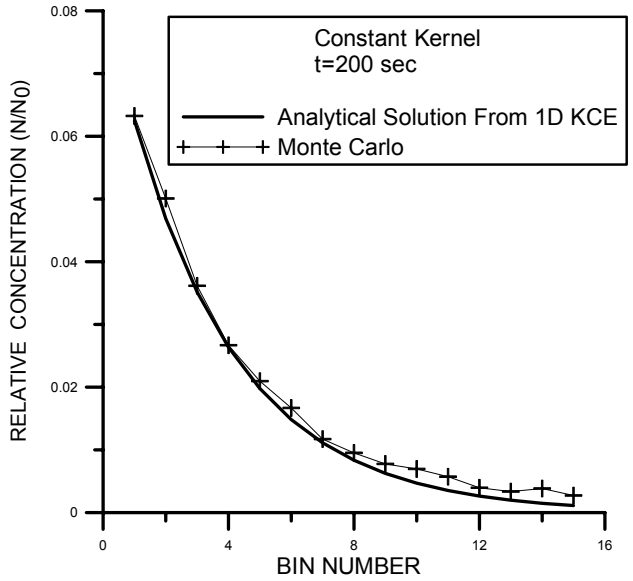

Fig. 9. Same as Fig. 8 , but for t=200.

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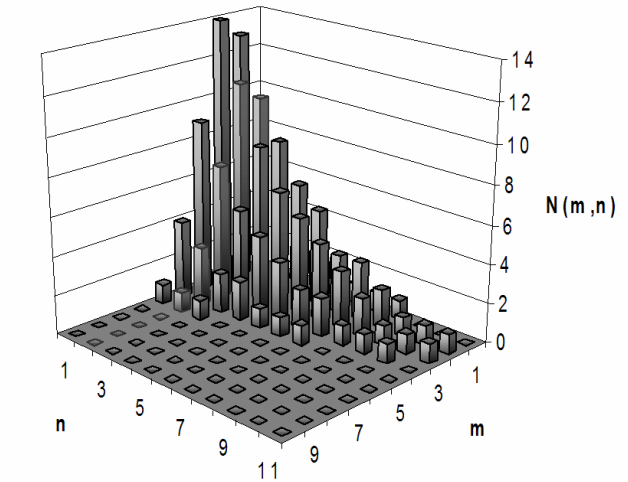

Fig. 10. Initial two-component spectrum $N(m, n, 0)$ with droplet concentration of 181 cm^{-3} and LWC 1.87 g/kg.

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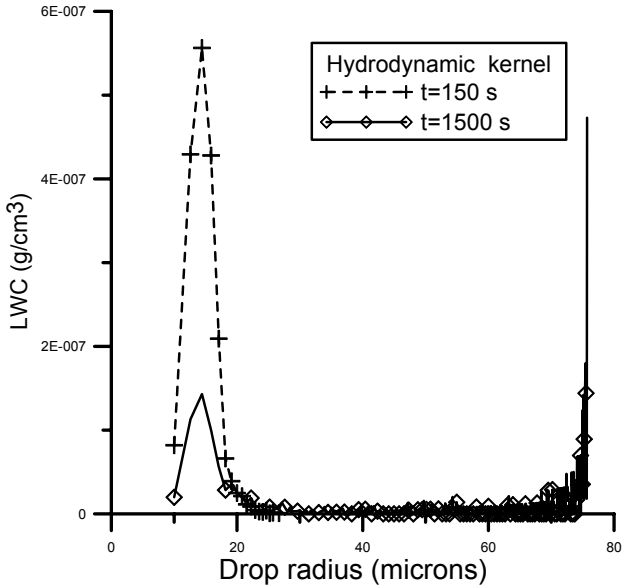

Fig. 11. Liquid water content (LWC, in g/kg) as a function of drop radius for the hydrodynamic coalescence kernel for t=150 s (dashed line) and t=1500 s (solid line with diamonds).

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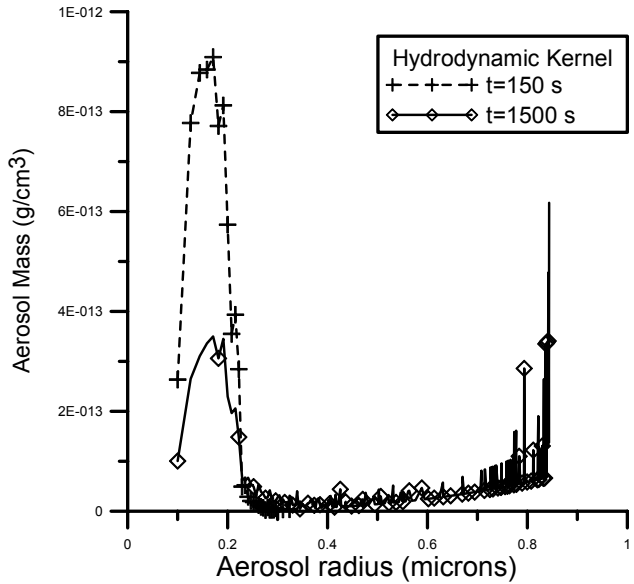

Fig. 12. Aerosol mass concentration (in g/cm^3) as a function of aerosol radius for the hydrodynamic kernel for t=150 s (dashed line) and t=1500 s (solid line with diamonds).

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