

# A Microscopic Aggregation Model of Droplet Dynamics in Warm Clouds

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A microscopic model of warm clouds involving input of water droplets, droplet-droplet aggregation, droplet breakup, and precipitation is presented. Numerical simulations and analytical arguments indicate that after the stage of growth and just before precipitation sets in, a warm cloud is characterized by a droplet-size distribution which follows from an inverse power law as a function of the droplet size. When precipitation is taken into account, the above distribution is transformed into a distribution decaying exponentially with the droplet size, in agreement with field observations. It is suggested that the initiation of rainfall in a precipitating warm cloud can be viewed as an instability triggered by the presence of a power-law distribution.

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**KEY WORDS:** Aggregation; cloud microphysics; power-law distributions; random walk.

## 1. INTRODUCTION

Clouds rank among the most complex and intriguing natural phenomena. A vast amount of literature is devoted to their formation and further evolution as well as to the conditions under which a cloud can cause precipitation.<sup>(1,2)</sup>

A cloud starts with the condensation of water vapor around condensation nuclei present in the upper levels of the earth's atmosphere. Typical nucleus sizes involved in this process of heterogeneous nucleation are of the order of micrometers or less. Homogeneous nucleation, while in principle possible, turns out to be not very important in the earth's atmosphere.

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With the continuous inflow of small droplets the cloud grows. As it ascends, its top may sometimes reach levels where the temperature is below the freezing point. Part of the liquid mass may then freeze and complex phenomena characteristic of aerosol clouds may occur. We do not consider such a possibility in the present paper, but focus entirely on "warm" clouds.

A stable, nonprecipitating, warm cloud is an assembly of water droplets with a density of the order of  $10^6$  droplets per  $\text{dm}^3$  of cloud. A droplet radius is on the average about  $r = 10 \mu\text{m}$  and typically beyond this value the number of droplets of a given size in the cloud decreases with the droplet size.<sup>(1)</sup> A common characteristic shape of droplet size distributions measured in different types of clouds corresponds to a probability density rising sharply from a low to a maximum value and then decreasing again for larger sizes, such that when plotted on a semilogarithmic scale, it exhibits a linear region in the range of large droplet sizes indicative of exponential decay. Other types of distributions are also observed, notably as a function of height, including bimodal ones.<sup>(1,2)</sup> Still one common characteristic in the stable nonprecipitating phase of clouds remains the short-tail, exponential-like nature of the droplet size distribution in the region of large sizes.<sup>(3)</sup>

In order that precipitation be initiated, a broadening of the droplet size distribution is clearly necessary to account for a substantial fraction of large drops falling under the effect of gravity. Typical raindrop sizes are of the order of  $100\text{--}1000 \mu\text{m}$ , with about 1 drop per  $\text{dm}^3$  of air. Field observations and laboratory experiments have shown that the drop size distribution of rain follows also on the average an exponential decay in the range of large drop sizes.<sup>(4)</sup>

The traditional approach to cloud modeling is based on a continuous formalism combining the equations of fluid dynamics together with the thermodynamic relations describing phase transformations.<sup>(2)</sup> Until now, within the frame of reference of such an approach, the mechanisms causing the broadening of the cloud drop distribution and thus controlling the embryonic precipitation in clouds have not received a satisfactory interpretation. For instance, some researchers attribute it to the presence of a significant concentration of giant aerosol particles,<sup>(5)</sup> while others regard it as a result of large-scale turbulent mixing and flow.<sup>(6)</sup> In the present paper a microscopic lattice model of a cloud is developed, which naturally leads to the broadening of the droplet size distribution. The model involves the input of small droplets, droplet-droplet aggregation, and in some cases the effect of spontaneous droplet breakup.<sup>(2)</sup> Any initial drop size distribution subjected to the combined effects of the above processes is transformed into a final drop size distribution which follows a power law in the large size range. When, in addition, precipitation is accounted for, through the

removal of cloud droplets, it is shown that the remaining droplets organize to form a stable cloud in which the droplet distribution exhibits a short tail in the same large size range.

The work is organized as follows. In Section 2 we describe the model and present some analytical results based on a mean-field description of a cloud in the growth phase (G phase) and in the mature, precipitation phase (P phase). Subsequently, we report, in Section 3, on the microscopic simulations performed in two dimensions for both cases considered in Section 2. The impact of the breakup mechanism on the steady state of the cloud during the G phase is also investigated. In the final Section 4, we compare our results with other theoretical and experimental findings and draw the main conclusions.

## 2. THE MODEL AND THE MEAN-FIELD ANALYSIS

Consider a volume of cubic shape within a developing cloud, located far from the cloud boundary and containing a large number of droplets,  $N$ . The droplets are placed on the sites of a regular cubic lattice and interact with each other through aggregation processes. Our objective will be to investigate, first, the growth process leading to a mature cloud and, second, the process leading eventually to precipitation. In both cases the central quantity to evaluate will be the drop-size distribution and, most particularly, its structural change as the cloud switches from the growth to the precipitation phase and back.

### 2.1. Growth Phase

At each given time a constant influx of stable small droplets placed randomly on the lattice sites is stipulated. The sizes of these droplets are supposed to exceed the critical nucleation radius<sup>(1)</sup> and to be distributed according to the probability distribution  $p(I)$ . Once on the lattice, droplets perform a complex motion due to such factors as molecular diffusion, turbulent mixing, etc. It appears reasonable to approximate this complex motion with a diffusive random walk, which for simplicity will be assumed independent of the droplet size. When droplets meet during a collision, it is assumed that they aggregate instantaneously, creating droplets of larger and larger sizes. Droplet splitting and loss are neglected at this stage; loss plays an important role during the precipitation phase and will be discussed in the next subsection, whereas the general effects of particle splitting will be considered in Section 3.

To proceed further, we make a mean-field assumption, known to be valid in space dimensions  $d \geq 2$ ,<sup>(7,8)</sup> and in the absence of anisotropy

induced, for instance, by gravity or other external fields. Within the framework of such an assumption the state of the system is described by the probability  $p(m, t)$  that at time  $t$  a water droplet of mass  $m$  will be located at some representative site. We also assume that  $p(m, t)$  evolves according to a Markovian process, whereby the probability to obtain a mass  $m$  at a certain site will only depend on the number of water droplets that were located on sites  $i, i = 1, \dots, r$  at time  $t - 1$  and subsequently jumped on the site under consideration during one time unit. In addition to the aggregating cloud droplets, one has to add the contribution of the input term. One may thus write

$$p(m, t) = p(I) \sum_{r=0}^N \binom{N}{r} \left(\frac{1}{N}\right)^r \left(1 - \frac{1}{N}\right)^{N-r} \prod_{i=0}^r p(m_i, t-1) \Big|_{\sum_{k=0}^r m_k + I = m} \quad (1)$$

Here  $N$  is the total number of lattice sites and the combinatorial factor stands for the various ways to choose  $r$  particular sites among the  $N$  available ones, each site having an *a priori* probability  $1/N$  to be chosen. The restriction  $m_1 + m_2 + \dots + m_r + I = m$  expresses the fact that the sum of the masses of the droplets emanating from these sites plus the mass of the incoming droplets must total the mass of the site considered. In writing Eq. (1), one has also implicitly assumed that the coalescence efficiency is equal to unity. Introducing the generating function  $Z(\rho, t)$  as the Fourier transform of  $p(m, t)$ ,

$$Z(\rho, t) = \int e^{-i\rho m} p(m, t) dm \quad (2)$$

one may transform Eq. (1) into the form

$$Z(\rho, t) = \int e^{-i\rho m} dm \times p(I) \sum_{r=0}^N \binom{N}{r} \left(\frac{1}{N}\right)^r \left(1 - \frac{1}{N}\right)^{N-r} \prod_{i=0}^r p(m_i, t-1) \Big|_{\sum_{k=0}^r m_k + I = m} \quad (3)$$

Using again Eq. (2) and introducing the time-independent input generating function  $\Phi(\rho)$  as

$$\Phi(\rho) = \int e^{-i\rho I} p(I) dI \quad (4)$$

one finally obtains

$$Z(\rho, t) = \Phi(\rho) \left(1 - \frac{1}{N}\right)^N \sum_{r=0}^N \binom{N}{r} \left(\frac{Z(\rho, t-1)}{N}\right)^r \quad (5)$$

In the limit of  $N \rightarrow \infty$ , Eq. (5) can be written in a closed form,

$$Z(\rho, t) = \Phi(\rho) e^{Z(\rho, t) - 1} \tag{6}$$

We now seek for a stationary solution of this equation. At first sight this seems paradoxical, since all loss mechanisms have been discarded at this stage. Still we remark that while growing, the dynamical system may attain a *statistically stationary distribution* of particle sizes.<sup>(7,8)</sup> This is precisely the quantity of interest in the present work. Assuming that such a distribution exists, we have in the long-time limit

$$Z(\rho) = \Phi(\rho) e^{Z(\rho) - 1} \tag{7}$$

Since we are interested in the tail of the steady-state mass distribution function  $p(m)$ , we seek solutions of this functional equation in the range of large droplet sizes or, from Eq. (2), of small Fourier variable  $\rho$ . Assuming that  $\Phi(\rho)$  is sufficiently short ranged so that  $\langle I \rangle \neq \infty$  (a case that includes, among others, Gaussian and exponential laws), we therefore write

$$\Phi(\rho) = 1 - i\langle I \rangle \rho + \dots \tag{8}$$

in lowest order of  $\rho$ . Furthermore, for any generating function corresponding to a continuous distribution<sup>(9)</sup> one may write the limiting form

$$Z(\rho) = 1 - c|\rho|^\alpha \tag{9}$$

where  $c$  is a constant number and  $\alpha$  is an exponent which remains to be determined. This entails that  $Z(\rho) - 1 \sim |\rho|^\alpha$  and can be considered as a small variable. By expanding the right-hand side of Eq. (7) in lowest order of  $\rho$ , we finally obtain an explicit value for the exponent  $\alpha$  and the generating function  $Z(\rho)$  as

$$Z(\rho) = 1 - c' e^{i\pi/4} \langle I \rangle^{1/2} |\rho|^{1/2} \tag{10}$$

where the imaginary constant factor indicates that the distribution to which the generating function belongs is one-sided and not symmetric around some mean value.<sup>(10)</sup> By taking the inverse Fourier transform of Eq. (10), we obtain<sup>(7,8,10)</sup>

$$p(m) \sim m^{-3/2} \tag{11}$$

showing that the steady-state mass distribution of large droplets in the growing phase follows an inverse power law. Notice that the existence of a well-behaved steady-state droplet size distribution provides an *a posteriori* justification of the working hypothesis that the cloud life cycle can be decomposed into two distinct phases.

## 2.2. Precipitation Phase

Equation (11) entails that the cloud produced during the growth phase is charged with many heavy droplets. This favors an instability, since large droplets start quitting the cloud due to gravity, thus giving rise to precipitation. When rain in the above sense is included in the cloud model, three phenomena take place simultaneously: input of particles, aggregation, and loss of droplets.

We can extend the mean-field calculation of Section 2.1 to incorporate precipitation in a very simple way. We assume that water droplets fall out of the system in a random manner, independent of their mass. This assumption is not entirely satisfactory, since it is expected that due to gravity, larger droplets are the ones that will eventually precipitate in a cloud. However, the random loss assumption will be adopted for simplicity and will give us a good idea of the effects of the precipitation mechanism and its influence on the evolution of the warm cloud.

We start again from the probability  $p(m, t)$  to find a particle of mass  $m$  at a particular time  $t$  in the cloud [see Eq. (1)]. We now need to take into account that in order to create a particle of mass  $m$  at a given site, the system has to spend a total mass equal to  $(1 + \lambda)m$ ,  $\lambda$  being the percentage of the droplets leaving the system,

$$p(m, t) = p(I) \sum_{r=0}^N \binom{N}{r} \left(\frac{1}{N}\right)^r \left(1 - \frac{1}{N}\right)^{N-r} \prod_{i=0}^r p(m_i, t-1) \Big|_{\sum_{k=0}^r m_k + I = m + \lambda m} \quad (12)$$

By Fourier transforming Eq. (12) and by following the same lines as in Section 2.1, we finally obtain the following relation for the droplet-size distribution in the long-time regime:

$$Z(\rho) = \Phi \left( \frac{\rho}{1 + \lambda} \right) \exp \left[ Z \left( \frac{\rho}{1 + \lambda} \right) - 1 \right] \quad (13)$$

If we assume again that the input distribution has a short tail and a mean  $\langle I \rangle \neq 0$  and  $\langle I \rangle \neq \infty$ , we may write

$$\Phi(\rho) = 1 - i \langle I \rangle \rho + \dots \quad (14)$$

and from Eq. (13)

$$Z(\rho) = \left( 1 - i \langle I \rangle \frac{\rho}{1 + \lambda} \right) \exp \left[ Z \left( \frac{\rho}{1 + \lambda} \right) - 1 \right] \quad (15)$$

Contrary to the growth phase,  $Z(\rho)$  can now be expanded in terms of simple powers of  $\rho$ ,

$$Z(\rho) = 1 - i\langle m \rangle \rho - \frac{1}{2}\langle m^2 \rangle \rho^2 + \dots \quad (16)$$

By substituting Eq. (16) into Eq. (13) and keeping the lowest order of  $\rho$ , we can find the average mass of the cloud droplets as

$$\langle m \rangle = \frac{\langle I \rangle}{\lambda} \quad (17)$$

We conclude that

$$Z(\rho) = 1 - i \frac{\langle I \rangle}{\lambda} \rho \quad (18)$$

to the lowest order in  $\rho$ . In other words, the corresponding distribution  $p(m)$  is short ranged with a finite mean, behaving qualitatively as an exponential in the limit of large size droplets. Notice that the mean mass of the droplets is proportional to the input and inversely proportional to the output ratio, as intuitively expected.

The physical meaning of the above result is that the random loss process narrows the size spectrum of the mass distribution so that at the steady state only a negligible number of large droplets remain in the cloud. As a result, precipitation stops and the cloud, if still subject to a continuous input of droplets, regains the growth phase.

A more realistic approach to account for precipitation would be to assume that the droplets fall with probability proportional to their mass, i.e., the larger droplets are more likely to fall due to gravity. We believe that this more realistic consideration will only enhance and accelerate the approach to the steady-state distribution, which is expected to become even narrower. Further work is needed to verify this conclusion.

### 3. NUMERICAL EXPERIMENTS

In this section we develop a cloud simulation algorithm describing the microscopic dynamics of the water droplets. We will again address separately: the G phase, during which the cloud grows because of a random input of small droplets and subsequently through a droplet-droplet aggregation mechanism; and the precipitation phase, during which, in addition to the above phenomena, loss of particles is also taken into account.

For computational reasons we limit ourselves to a two-dimensional lattice, which may be thought of as a horizontal section of the cubic volume introduced in Section 2. As a consequence the effect of gravity will not be considered.

### 3.1. Growth Phase

We start with the lattice of  $N$  sites in which every site is initially occupied by a water droplet of mass  $m = 1$ . At every time step, a number of the order of  $N/3$  water droplets of the smallest size, representing the continuous inflow of mass from the ambient medium, reach the lattice sites and coagulate with the already existing droplets. Moreover, in order to model turbulent phenomena taking place inside the cloud, each droplet is allowed to move randomly to one of its nearest neighbors, independent of its size. Aggregation occurs if two or more such particles happen to meet on the same lattice site, at the same time, producing in this way a new droplet which carries now the sum of the masses of the incoming particles. Finally, the boundary conditions used are periodic.

Following the above algorithm we have first evolved two-dimensional clouds of sizes  $20 \times 20$  to  $200 \times 200$  in the absence of breakup until a steady-state probability distribution is reached. In Fig. 1 we have plotted in a double logarithmic scale the cumulative drop size distribution  $P(m)$  (dots) as well as the probability density function  $p(m)$  (crosses) as a function of the droplet size  $m$  obtained for a  $100 \times 100$  lattice after 15,000 iterations.

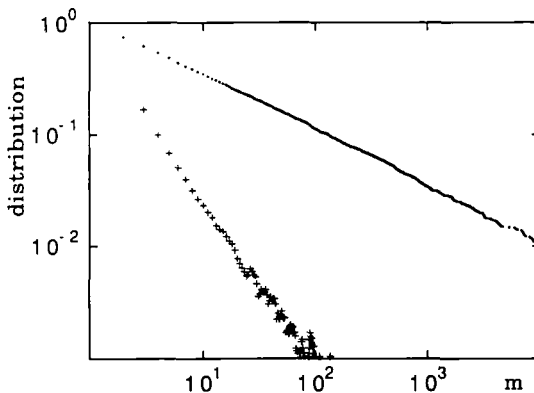


Fig. 1. Asymptotic form of the cumulative probability distribution  $P(m)$  (dots), and the probability density  $p(m)$  (crosses) as a function of the droplet size  $m$ , obtained after 15,000 time units from a  $100 \times 100$  lattice, simulating a horizontal section of a warm cloud in the G phase



We see that the cumulative droplet-size distribution follows a power law with an exponent  $-0.5$ ,  $P(m) \sim m^{-0.5}$ . Since the cumulative drop-size distribution function  $P(m)$  is the integral of the drop-size probability density function  $p(m)$ , we conclude that in the limit of large mass<sup>(8)</sup>

$$p(m) \sim m^{-1.5} \quad (19)$$

which is in quantitative agreement with the analytical results based in the mean-field description of Section 2. This agreement is of no surprise, since in random walk and aggregation-related processes it is known that for dimensions  $d \geq 2$  the mean-field description is expected to hold.<sup>(11)</sup>

Notice that the initial distribution does not affect the numerical results, since it is soon forgotten due to the rearrangements performed by the aggregation and input processes. In addition, the steady-state results are independent of the input rate of droplets and the lattice size, provided that the latter exceeds a threshold below which boundary effects are dominant. A more realistic input model should involve an input of the form of a Gaussian-like distribution of droplets centered at reasonably small sizes. In this case, one expects that the part of the probability distribution corresponding to the small size drops of Fig. 1 will be replaced by a curve rising sharply from the origin to a maximum value, corresponding roughly to the maximum of the input term, and then decreasing according to Eq. (11).

In Fig. 2 we show a snapshot of a cloud in the G phase, characterized by a drop-size distribution which has achieved the power-law behavior of

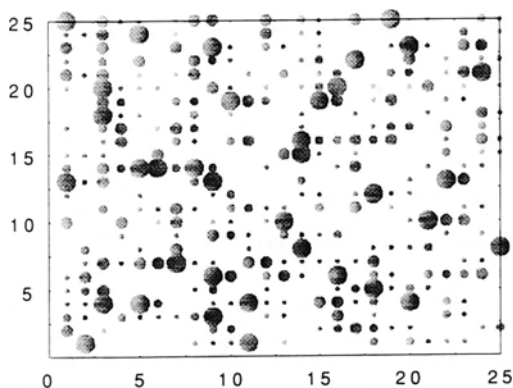


Fig. 2. Snapshot of a  $25 \times 25$  portion of a  $200 \times 200$  lattice in which the asymptotic distribution represented in Fig. 1 has been attained. The four classes of drops, small, intermediate, moderate, and large represent, respectively, 40, 30, 20, and 10% of the total probability mass.

Eq. (11). The drops have been subdivided into four classes according to their size range. The ranges of small, intermediate, moderate, and large drops are chosen to represent, respectively, 40, 30, 20, and 10% of the total probability mass. The size of the dots in Fig. 2 follows the above subdivision, while empty sites have not been marked. For clarity only a  $25 \times 25$  portion of the total  $200 \times 200$  lattice is shown. We notice that the distribution is highly irregular, owing presumably to the presence of inhomogeneous fluctuations. A box counting algorithm<sup>(12)</sup> of this instantaneous profile has been used to estimate the dimensionality  $D_0$  of the large droplet set as well as of the entire droplet set. In both cases the dimension turns out to be  $D_0 \simeq 1.9$ . Simulations of larger size clouds are necessary before drawing definitive conclusions about the fractal character of  $D_0$ .

As we mentioned in the Introduction, besides diffusion and aggregation, other processes also participate in the formation of the droplet-size distribution. In particular, the droplet breakup occupies a reasonable part of the cloud literature.<sup>(2)</sup> Droplets can break either spontaneously or during collisions in all phases of the life of a cloud. In the following, we include a simple breakup mechanism during the G phase, in order to get a first estimation on the modifications of the droplet size distribution due to breakup.

To take this effect into account, one needs to modify slightly the growth algorithm by adding a step during which the droplets can break with a certain probability. To simplify the simulations, we assume that a droplet of a given initial size  $m$  can break into two droplets of random sizes  $m'$  and  $m''$  such that  $m' + m'' = m$ . One of the two resulting droplets

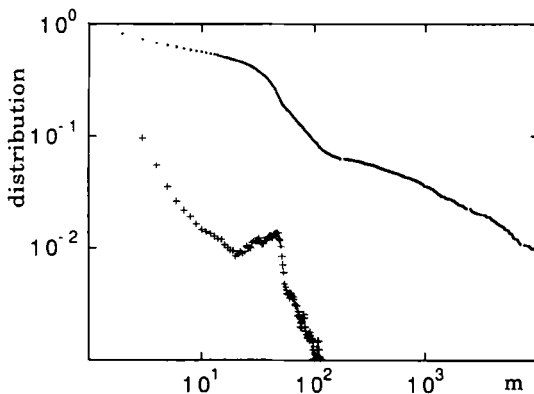


Fig. 3. As in Fig. 1, but in the presence of droplet breakup with  $m_1 = 50$ ,  $m_2 = 200$ , and breakup probability  $pbreak = 0.5$ .

remains on the original site, while the other chooses at random to aggregate with one of the four nearest neighbors.

In Fig. 3 we show the cumulative probability distribution (dots) and the probability density (crosses) when breakup is considered in the G phase. Since this mechanism is strongly dependent on the masses<sup>(13)</sup> of the droplets we have adopted the reasonable prescription that only masses in a range, say, between  $m_1$  and  $m_2$  may lead to breakup. We observe in the cumulative distribution a qualitatively new feature in the form of an inflection point, indicative of a maximum in the drop-size probability distribution.

We suggest that this result constitutes the signature of the effects of the breakup mechanism in the features of the steady state. It is therefore reasonable to conjecture that the maximum observed for intermediate drop sizes in the probability distribution in real clouds is a consequence of the breakup mechanism.

### 3.2. Precipitation Phase

A cloud that has accumulated large quantities of water vapor during the G phase and has achieved a power-law drop-size distribution is a "heavy," unstable cloud containing many droplets of large size. It is reasonable to expect that this cloud will start to precipitate. We extend the previous algorithm to include precipitation as follows:

- (a) Start again with a two-dimensional lattice and on every site of the lattice put a particle (droplet) of random size (mass). The initial mass distribution is not important, it will be forgotten for long times due to the processes of input, aggregation, and precipitation.
- (b) At every time step introduce randomly vapor droplets of size 1, on every lattice site with a certain probability "*pinput*," as in the case of the G phase. In these experiments we use  $pinput = 1/3$ .
- (c) Move randomly all the particles to one of their nearest-neighbor sites initiating the aggregation phenomena as in the G phase.
- (d) Finally, with a certain probability "*prain*" remove particles off the lattice, as rain. Here we use  $prain = 1/3-1/5$ .

To simplify our model, we have neglected the effect of droplet breakup during the P phase. The above mechanism for the production of rain in which every particle can drop out of the cloud as rain is a very simplistic

one. In reality, as pointed out in Section 2, we should expect that only the largest droplets will produce rain.

Using the above algorithm we evolved two-dimensional clouds of sizes  $20 \times 20$  to  $200 \times 200$  sites, for up to 15,000 time steps. In Fig. 4 we show the drop-size distribution as a function of the droplet size.

In a simple logarithmic scale the data follow, for large droplet sizes, a straight line, which indicates an exponential decay behavior. This exponential behavior agrees with the mean-field result on aggregation with input and loss described in Section 2.

In Fig. 5a we show the drop-size cumulative probability distribution of precipitation just underneath the cloud as a function of the droplet mass. A clear-cut exponential law appears, for large  $m$ , in agreement with the analytical results of Section 2. On the other hand, traditionally, the probability density obtained by field observation data can be fitted grossly by an exponential law as a function of the droplet diameter  $D$ .<sup>(1,2)</sup> As seen from Fig. 5b, there is a range of radii in which the distribution as a function of  $D$  can also be fitted reasonably well by an exponential law. The effective exponent  $A$  turns out to be  $A \sim -1.8$ . Expressing  $A$  in terms of the rainfall rate  $R$  from the empirical Marschal–Palmer law<sup>(1,2,24)</sup>

$$|A| = 4.1R^{-0.21} \quad (20)$$

one obtains then information on the important parameter  $R$ . At this stage quantitative results cannot be claimed, as no space and time scales have been introduced in the lattice model.

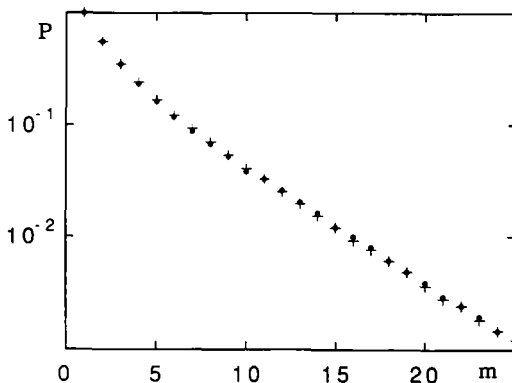


Fig. 4. Asymptotic form of the cumulative probability distribution  $P(m)$  for a cloud in its P phase after 7500 (dots) and 15,000 (crosses) time units.

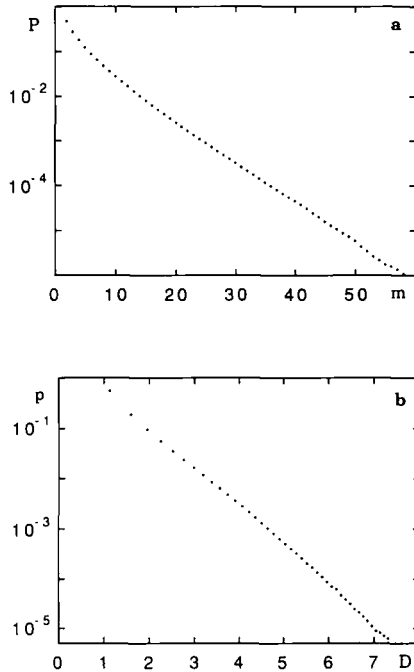


Fig. 5. (a) The cumulative probability distribution  $P$  of the precipitating droplets as a function of the droplet mass  $m$ , (b) the probability density  $p$  as a function of the droplet diameter  $D$ , resulting from a  $100 \times 100$  cloud lattice after 15,000 time units.

#### 4. CONCLUSIONS

We have studied theoretically in the mean-field limit and numerically in two dimensions the evolution of a warm cloud, modeled as an ensemble of aggregating water droplets. We have shown that the combined effects of aggregation and water input create a “heavy” cloud containing many droplets of large mass described by a drop-size distribution in the form of a power law  $p(m) \sim m^{-1.5}$ . When a cloud is so heavily charged, precipitation is initiated. We have shown that the combined effects of input, aggregation, and loss lead to a “light” cloud with only few droplets of large mass. In fact the drop-size distribution exhibits now an exponential decay.

It is tempting to envisage a cloud as a collection of water droplets oscillating between power-law behavior and exponential decay. If we keep the input and loss fluxes constant, these oscillations are expected to be regular. However, in nature the input of vapor droplets does not have a constant rate, but varies according to the weather conditions, the season, the

availability of water vapor, etc. In other words, the cloud oscillations that one can observe in nature are of aperiodic nature rather than periodic ones.

Actually, one may arrive at a unified formulation of the two phases in the life of a cloud by considering that the parameter  $\lambda$  varies in a continuous range. When  $\lambda$  is finite, loss is present, the average droplet mass  $\langle m \rangle = \langle I \rangle / \lambda$  is finite, and the probability distribution reduces, in the range  $m \gg 1$ , to

$$p(m) \sim e^{-m/\langle m \rangle}, \quad m \gg 1 \quad (21)$$

As  $\lambda$  decreases, the average droplet mass increases and the exponential distribution becomes long-ranged in the limit  $\lambda \rightarrow 0$ ,  $\langle m \rangle \rightarrow \infty$ , where the probability distribution is unnormalizable. This effect signals the failure of the procedure leading to Eq. (16). Slowly decaying terms become then dominant, thus leading to the inverse power-law terms found in the G phase. This change in behavior is analogous to what happens in a phase transition. In this respect  $\lambda$  may be considered as the distance from criticality, whereas  $\langle m \rangle$  plays the role of the order parameter.

In the range of intermediate values of droplet size we have shown that the spontaneous breakup of the cloud droplets gives rise to a hump in the probability distribution. Keeping in mind that for very small sizes an additional sharp maximum is likely to occur due to the nature of the input distribution, it seems legitimate to assert that this provides a plausible mechanism for the bimodality observed in certain types of clouds.

A different growth and coalescence algorithm has recently been developed by Meakin.<sup>(15)</sup> The model involves input of particles, local growth aggregates, and loss. In the present work in addition to growth and coalescence, specific mechanisms relevant to the life cycle of warm clouds have been incorporated, such as droplet motion, breakup, and loss by precipitation.

All our simulations have been performed in two dimensions. Clearly three-dimensional simulations are desirable in order to model more closely the dynamics of a real cloud. However, the static behavior and many dynamical properties of random walks and aggregation are not very different in two and three dimensions. One therefore expects that the asymptotic behavior of the two-dimensional cloud will also persist in three dimensions, since  $d=2$  is the upper critical dimension for these types of models. Consequently, the same type of instability and initiation of precipitation is expected in three dimensions.

In a future work a more elaborate analysis is planned, taking into account various types of droplet breakup mechanisms, alternative loss mechanisms favoring heavy particles in the process of precipitation, as well as investigations on the role of the vertical dimension.

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