Notes on the state-of-the-art numerical modeling of cloud microphysics

A. Khain\textsuperscript{a,\ast}, M. Ovtchinnikov\textsuperscript{b}, M. Pinsky\textsuperscript{a}, A. Pokrovsky\textsuperscript{a}, H. Krugliak\textsuperscript{a}

\textsuperscript{a} The Institute of Earth Sciences, The Hebrew University of Jerusalem, Givat Ram Campus, Jerusalem, 91904 Israel

\textsuperscript{b} CIMMS, University of Oklahoma, Norman, OK, USA

Received 23 February 2000; received in revised form 3 July 2000; accepted 31 August 2000

Abstract

Despite significant advances in cloud physics, many problems exist in the state-of-the-art microphysical cloud modeling. The progress is hampered by (1) many remaining gaps and uncertainties in our knowledge of cloud microphysics and (2) limitations of numerical approaches in representing some of known microphysical processes.

In this paper, we attempt to give an assessment of several important problems of warm and ice microphysics and model limitations and identify areas where improvements are most urgently needed. Because of the complexity and broadness of the subject, the review does not offer an exhausted analysis of the field or provide solutions for all discussed problems. We are concerned with the spectral (bin) microphysical approach, which does not restrict the particle size spectra to any particular shape and, therefore, claims to reproduce formation of size spectra of cloud particles. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Cloud microphysics; Precipitation formation; Droplet spectrum formation; Numerical modeling of cloud processes

1. Introduction

Clouds play a crucial role in the dynamics and thermodynamics of the atmosphere. Precipitation from clouds is of vital importance for human life and activity. Latent heat
release is one of the main energy sources for atmospheric phenomena of different spatial scales ranging from individual clouds, to mesoscale systems (e.g., squall lines and tropical cyclones), and the global circulation. Cloud radiative properties largely control the Earth’s climate and affect possible climatic changes. The rate of latent heat release, processes of precipitation formation, and radiative properties of clouds are affected by microphysical processes of formation, growth and interaction of drops and ice particles. Many of these processes are not well understood. Observations, though greatly improved in quality in recent years, have limitations that circumscribe our ability to study microphysical processes directly in clouds. Laboratory experiments give us an important insight into individual processes in a controlled environment, which is not necessarily representative of the cloud conditions. Theoretical studies encounter their own difficulties. Systems of equations describing all interacting physical processes in a cloud are so complex that analytical solutions are available only for highly idealized cases. Theoretical studies are related usually to some specific problems such as calculation of drop–drop, drop–ice and ice–ice collision efficiencies, simulation of diffusion growth of droplets in case of prescribed dynamics, etc. Considering the above, numerical modeling is emerging as a potentially invaluable tool in studying cloud processes under both natural and artificial cloud seeding conditions.

Despite significant advances in cloud physics, many problems exist in the state-of-the-art microphysical cloud modeling. The progress is hampered, in our opinion, by (1) many remaining gaps and uncertainties in our knowledge of cloud microphysics and (2) limitations of numerical approaches in representing some of known microphysical processes.

In this paper, we attempt to give an assessment of several important microphysical problems and model limitations and identify areas where improvements are most urgently needed. Because of the complexity and broadness of the subject, the review does not offer an exhausted analysis of the field or provide solutions for all discussed problems.

All cloud models can be separated into two broad groups, according to the microphysical representation used. Models of the first type utilize the so-called bulk-parameterization of microphysics. Models of the second kind are based on spectral (bin) approach to the description of microphysical processes. The bulk microphysical parameterization schemes are most commonly used in mesoscale models such as, for example, GCEM (Simpson and Tao, 1993), RAMS (Pielke et al., 1992; Walko et al., 1995) and MM5 (Reisner et al., 1998). In these schemes, all microphysical processes are described in terms of integral parameters, such as mass contents and, in some cases, number concentrations of a few types of cloud and precipitation particles. The relatively small number of the integral parameters makes the bulk parameterization schemes computationally efficient. In spectral (bin) microphysical models (e.g., Hall, 1980; Kogan et al., 1984; Flossman et al., 1985; Kogan, 1991; Khvorostyanov et al., 1989; Khain and Sednev, 1995, 1996; Khain et al., 1999a; Stevens et al., 1996; Reisin et al., 1996; Bott, 1997; Levin et al., 1998; Ovtchinnikov and Kogan, 2000) each type of cloud hydrometeors (e.g., water drops in warm clouds, and drops, ice crystals, aggregates, graupel and hail in mixed-phase clouds) is described using size (mass) distribution functions containing several tens of bins of masses. In contrast to the bulk parameterization
schemes, the shapes of the size distributions in spectral models are not determined a priori, but are calculated in the course of the model integration.

In this review, we are concerned with the latter (spectral) approach, which does not restrict the particle size spectra to any particular shape and, therefore, is able to reproduce cloud processes in a more realistic way. The term “explicit microphysics” has been ambiguously used in literature to describe both particle size resolving models as opposed to bulk parameterizations (e.g., Kogan, 1991), and bulk microphysical parameterizations in mesoscale models that “explicitly” predict cloud and rain water as well as ice water content for various types of ice particles as opposed to models which diagnostically determine these parameters (e.g., Reisner et al., 1998). Because of this ambiguity, we will avoid using the term “explicit” in this paper.

In Sections 2–4, the theoretical problems and technical assessment in modeling liquid and ice phase microphysics are discussed. They are related mostly to our gaps in understanding of certain physical processes and can only be solved by combining results from observational, laboratory, theoretical, and modeling studies. In Section 5, we present an example of model calculations indicating the importance of cloud aerosol interaction, or initial stage of cloud development on all sequent cloud life. Conclusions can be found in Section 6.

2. Warm rain microphysics: basic processes

Microphysical processes that are not related to ice formation are often referred to as warm rain microphysics.

2.1. Condensation growth

The growth rate of the atmospheric particles containing soluble parts is described by the equation of condensation (or diffusion) growth caused by the diffusion of water molecules to the particle (Pruppacher and Klett, 1997):

\[
\frac{dr}{dt} = \frac{1}{F} \left( S - \frac{A}{r} + \frac{Br_N}{r^3 - r_N^3} \right)
\]

(2.1)

where \( r \) is the radius of wet particle, \( r_N \) is the radius of dry soluble fraction of the particle, \( S \) is the value of supersaturation in an air parcel with respect to water, \( F \) is the coefficient depending on thermodynamics parameters such as temperature, thermal conductivity of air, etc. The second term on the right hand side is related to particle curvature, \( A = 2\sigma/\rho_N R T \), where \( \sigma \) is the surface tension of the droplet. The “solution” term \( B = i\rho_N M_w/M_s \), where \( i \) is the number of ions for each solvent molecule and equal to 2, \( \rho_N \) is the density of dry AP, \( M_i \) and \( M_w \) are the molecular masses of the solvent and water, respectively.

The particle growth by condensation begins below cloud base under \( S < 0 \) and continues in clouds under supersaturation conditions \( (S > 0) \), giving rise to the droplet formation.
For physical reasons stated below, the process of condensation growth of particles is usually separated into three stages:

(a) growth of aerosol particles to their equilibrium size;
(b) nucleation of droplets;
(c) diffusion growth of droplets.

2.1.1. Droplet nucleation and the formation of spectrum of newly nucleated droplets

2.1.1.1. Theory and observations. For each \( r_N \), one can derive an equilibrium supersaturation \( S_{eq}(r) \) as a function of wet particle radius \( r \). \( S_{eq}(r) \) corresponds to the condition when \( dr/dr = 0 \). Particles of radius \( r \) grow if \( S > S_{eq}(r) \) and evaporate if \( S < S_{eq}(r) \).

Dependencies \( S_{eq}(r) \), called Köhler curves (Rogers and Yau, 1989; Pruppacher and Klett, 1997), are determined mainly by the mass of dry soluble fraction of particles as illustrated in Fig. 1. Each curve corresponds to a certain value of \( r_N \). The Köhler curves shift down with an increase of \( r_N \), so that in the figure \( r_{N1} < r_{N2} < r_{N3} \).

The theory of the nucleation process is described in detail by Mazin and Shmeter (1983), Rogers and Yau (1989), and Pruppacher and Klett (1997) and is not repeated here. To illustrate the theory let us consider three cases.

(a) The line \( S = const \) crosses the Köhler curve in two points a and b (Fig. 1, curve 1). Corresponding size of particles will be \( r_a \) and \( r_b \). For particles with \( r < r_a \), \( S > S_{eq} \), so they grow tending to \( r = r_a \). For particles with \( r_a < r < r_b \), \( S < S_{eq} \) and these particles will partially evaporate until the size \( r = r_s \) is reached. Thus, point \( r = r_s \) is the stable

![Fig. 1. Dependencies of equilibrium supersaturation \( S_{eq} \) on radius \( r \) of wet particle (Köhler curves) (see text for details).](image-url)
point. For particles with \( r > r_p \), \( S > S_{eq} \) and these particles can grow without any limit. Thus, point \( r = r_p \) is the point of the unstable equilibrium with respect to perturbations on the right (larger radii). Particles with radii \( r > r_p \) are referred to as cloud droplets, while particles with radii \( r < r_p \) are referred to as non-activated (or wetted) aerosol particles (hereafter, AP). They tend to reach their equilibrium size \( r = r_e \). Thus, to become droplets, particles must be greater than \( r_p \) in radius. Otherwise, the particles remain to be non-activated AP.

(b) The line \( S = \text{const} \) touches the Köhler curve in one point (Fig. 1, curve 2). In this case \( S > S_{eq} \) for all particle sizes \( r \) except one point \( b' \), corresponding to \( r = r_{crit} \). The point \( b' \) is the point of the unstable equilibrium. The dry radius of aerosol particle \( r_N \) corresponding to curve 2 can be referred to as a critical size of dry aerosol particle under a given supersaturation \( S \).

(c) For particles with \( r_p > r_{NCrit} \), \( S > S_{eq} \) for all \( r \) (curve 3, Fig. 1), and they grow within the whole range of radii \( r \). Dry aerosol nuclei with radii \( r_p > r_{NCrit} \) are referred to as cloud condensation nuclei (CCN). Corresponding wet particles are referred as newly nucleated droplets (nucleation process).

Most particles reach their equilibrium size during a very short period of time (fraction of a second) (Kogan et al., 1984; Kogan, 1991). Thus, at \( S > 0 \) atmospheric particles can be separated into two main groups: growing cloud droplets and wetted non-activated AP, most of which are in the equilibrium with the environment. In case \( S < 0 \), only non-activated AP exist under stationary conditions.

2.1.1.2. Parameterization of the nucleation process. The size distribution and chemical composition of CCN together with the value of supersaturation determine the size distribution of newly nucleated droplets. Modelers use several approaches to parameterization of the nucleation process in cloud models.

According to the first approach, the concentration \( N \) of activated CCN (or concentration of nucleated droplets) is represented by an empirical dependence \( N(S) \). This dependence is often represented by formula (Pruppacher and Klett, 1997):

\[
N = N_0 S^k
\]  

(2.2)

where \( S \) is supersaturation (%), \( N_0 \) and \( k \) are constants determining the concentration of newly nucleated droplets. Expression (2.2) is also known as Twomey’s formula. Parameters \( N_0 \) and \( k \) (slope parameter in log–log scale), as well as the range of \( S \) within which formula (2.2), is valid, depend on size distribution and the value of soluble fraction of AP in a given air mass. These parameters vary in different geographic regions, over continents and oceans, urban and rural zones. According to the observational data presented by Pruppacher and Klett (1997, p. 289), \( 50 \text{ cm}^{-3} < N_0 < 250 \text{ cm}^{-3} \) and \( 0.3 < k < 0.6 \) for maritime clouds and \( 600 \text{ cm}^{-3} < N_0 < 5000 \text{ cm}^{-3} \) and \( 0.5 < k < 0.9 \) for continental clouds. According to the data (see Fig. 9.1 in Pruppacher and Klett, 1997), formula (2.2) is valid within a wide range \( 0.1% < S < 8% \). Note that the dependence (2.2) is determined under laboratory conditions using a Particle Measurement System (e.g., PMS FSSP-300 and PCASP-100X, ASASP-100X probes, Kim and Boatman, 1990a,b; Strapp et al., 1992), in which temperature of an air probe under investigation can be decreased (and supersaturation increased) within the range available.
in a given CCN counter. As supersaturation in real clouds is below several percent, formula (2.2), being valid within the range \(0.1\% < S < 8\%\), provides information about the smallest AP, which could be activated in clouds. As stressed by Cooper et al. (1997), the assumed distribution (2.2) must be modified for the largest particles because otherwise for soluble particles (2.2) gives a total CCN mass that is infinite for any \(k < 2\). Measurements carried out by Alofs and Liu (1981) suggest that a change in the slope occurs at a supersaturation of about 0.05\%. They report that the slope parameter \(k\) is about 3.85 for supersaturations lower than this. Cooper et al. (1997) note, however, that there is also extensive evidence for a prevalent “Junge” distribution of particles with sizes of about 0.1–1.0 \(\mu\)m, which for soluble particles would require \(k = 2\).

Hudson (1984, 1993), and Hudson and Frisbie (1991) presented the results of CCN measurements in extreme maritime cases. According to their data, a constant slope (parameter \(k\)) takes place only for \(S < 0.2\%\). At higher values of \(S\) the rate of new droplet generation rapidly decreases. No increase in CCN concentration was found for \(S > 0.6\%\). The decrease in the slope at higher supersaturations indicates that the amount of the smallest AP in the probes was so small that even a remarkable increase in \(S\) results in no nucleation of new droplets. Note that the data presented by Pruppacher and Klett (1997, Fig. 9.1–9.2) does not indicate the existence of such sharp reduction of CCN concentration with a decrease in CCN size. Note that these discrepancies in the behavior of CCN concentration with the increase in \(S\) above 0.2–0.3\% for maritime clouds are related to a very important problem, namely, to what extent supersaturation can grow in maritime clouds. In case of a limited number of small CCN, supersaturation can grow significantly accelerating droplet growth by condensation. In case, the concentration of small CCN is significant, the condensation growth will be slowed by the new droplet nucleation. Based on Hudson’s data, Cohard et al. (1998) stressed that “a more universal activation spectrum should exhibit a concave curvature in log–log scale (or in other words, a decrease of the \(k\) coefficient)” due to the limited availability of condensation nuclei as \(S\) is increased. So using Twomey’s original formula can induce important errors in predicting the nucleated cloud droplet number concentration unless the \(N_0\) and \(k\) parameters are “adjusted” to simulate a specific nucleation regime. Cohard et al. (1998) generalized Twomey’s approach providing modification of formula (2.2).

The data on height dependence of CCN concentration are very limited. It is widely accepted that CCN concentration decreases with height (Mazin et al., 1989). Exponential decrease is usually assumed: \(N_0 = N_0(z = 0) \exp\left(z/z_0\right)\), where \(z_0 = 1.5\) km. Note, however, that AP of different sizes can have different origins, so that the concentration of small particles decreases with height (if any) slower than that of the largest ones.

The knowledge of height dependence of CCN concentration becomes very important in case droplet spectrum broadening is attributed to the activation of CCN by dry air entrainment and mixing with cloud surroundings at significant distances above the cloud base.

Expression (2.2) is often used to determine the total concentration of newly nucleated droplets (e.g., Reisner et al., 1998). The mean radius of nucleated droplets or the size distribution of newly nucleated droplets is often a priori preset as being typical of a certain geographical region. For example, Reisin et al. (1996) assume initial droplet size...
distribution of gamma or exponential form centered at 11- and 5-μm radii for maritime and continental clouds, respectively. These spectra, especially for maritime clouds, contain relatively large droplets that may lead to drop collisions early in a simulation.

In more detailed approaches, the budget of CCN is calculated. CCN are transported by convective motions and activated in case when a certain value $S$ is attained. In this approach CCN are described using size the distribution function $f_N(x,r_0)$ depending of $N$. CCN size and spatial coordinates e.g., Respondek et al., 1995. The distributions typical of maritime, continental or “mixed” conditions are usually specified at the beginning of simulation. The process of CCN nucleation is usually described as follows. Assuming equilibrium conditions, the derivative $dS/dr_{crit}$ can be obtained from (2.1) under the equilibrium assumption $dr/dt = 0$.

The minimum (critical) values of the radius of nucleated droplet $r_{crit}$ and the radius of activated CCN particle $r_{Ncrit}$ can be expressed as:

$$r_{crit} = \frac{2A}{3} \frac{1}{S}$$

(2.3)

and

$$r_{Ncrit} = \frac{A}{3} \left( \frac{4}{B S^2} \right)^{1/3}$$

(2.4)

Using the values of supersaturation calculated in a cloud model, the values of $r_{crit}$ and $r_{Ncrit}$ can be determined using (2.3) and (2.4) and the size distribution of newly nucleated droplets can be calculated (e.g., Flossman et al., 1985; Kogan, 1991; Wurzler et al., 1995; Khain and Sednev, 1996; Yin et al., 2000). These calculations should be carried out with caution because the equilibrium sizes of droplets formed on giant and ultragiant CCN can be as large as several tens of microns in radius. For example, a NaCl particle of 10 μm in diameter has a 50-μm equilibrium diameter under 99% relative humidity. However, it takes a long time (sometimes days) for such a particle to reach such an equilibrium size. In case the equilibrium is assumed for all droplets, the initial droplet spectrum will be artificially broadened and collisions will become effective just above the cloud base.

Detailed calculations conducted by Ivanova et al. (1977) showed that if the radius of a soluble part of an aerosol particle $r_y < 0.03 \mu m$, the mass of water condensing on such CCN can be calculated under the equilibrium assumption (the Köhler equation), while for $r_y > 0.03 \mu m$, the mass of water condensing on these CCN at zero supersaturation can be calculated as $m_w = K(4/3)\pi r_y^3 p_w$, where $3 < K < 8$. This expression is also recommended by Kogan et al. (1984). Under the assumption that CCN particles are fully soluble, the latter expression determines the size distributions of nucleated droplets.

A similar approach has been used recently by Khain et al. (1999a) and Yin et al. (2000). This approach provides narrowing of the initial droplet spectra and prevents the unrealistically fast raindrop formation.

To evaluate the time period during which AP are located below the cloud base and, consequently, the size of AP at the cloud base, the vertical velocity below the cloud base should be taken into account (Kogan, 1991).
Note that the empirical formula (2.2), as well as any empirical formula \(N(S)\) within the range of its validity, determines the size distribution of CCN (APs with size \(r_N > r_{\text{crit}}\)) and nucleated droplets (Mazin and Shmeter, 1983; Emde and Wacker, 1993) with respect to supersaturation \(S\). Indeed, each value of \(r_{\text{crit}}(S)\) corresponds to a certain value of supersaturation \(S\). Using (2.2) one can write the following expression for aerosol size distribution as:

\[
\frac{dN}{dr_{\text{crit}}} = N_0 k S^{k-1} \frac{dS}{dr_{\text{crit}}}. \tag{2.5}
\]

After simple rearrangements, size distributions of CCN and nucleated droplets can be rewritten as follows:

\[
\frac{dN}{d\ln r_{\text{crit}}} = \frac{dN}{d\ln r_{\text{crit}}} = \frac{3}{2} N_0 k S^k \tag{2.6}
\]

where \(r_{\text{crit}}\) and \(r_{\text{crit}}\) can be determined using (2.3) and (2.4).

In case (2.2) is valid for \(S > 0.1\%\), which corresponds to \(r_N < 0.07\ \mu\text{m}\), expression (2.6) provides size distribution for small CCN only. However, in many cases when particles with a soluble fraction \(r_N < 0.07\ \mu\text{m}\) represent major fraction of CCN concentration, formulas (2.2) and (2.6) provide a reasonably good evaluation of the concentration of nucleated droplets and the values of supersaturation (Mazin and Shmeter, 1983), as well as size distribution of nucleated droplets with radii up to 0.4–0.5 \(\mu\text{m}\). Note that formulas similar to (2.6) can be used for the reconstruction of CCN size distribution from any observed dependencies \(N(S)\). As the expression \(N(S)\) (within the range of its validity) determines the size distribution of nucleated droplets, the latter cannot be preset independently.

In a series of studies (Ghan et al., 1993, 1995; Bedos and Rosset, 1996; Abdul-Razak et al., 1998), a method of determination of droplet concentration in clouds by nucleation is proposed. The idea of the method is based on the classical theory predicting the formation of supersaturation maximum within 10–30 m above the cloud base. The rate of supersaturation change in the liquid-phase cloud parcel is expressed by equation (cf. Squires, 1952; Squires, 1958; Politovich and Cooper, 1988; Pruppacher and Klett, 1997):

\[
\frac{dS}{dt} = A_1 W - A_2 \frac{dq_L}{dt}, \tag{2.7}
\]

where \(W\) is the updraft speed, \(q_L\) is the molar mixing ratio of liquid water in the air, and the coefficients \(A_1\) and \(A_2\) depend on thermodynamic parameters (temperature, latent heat of vaporization, air viscosity, etc.). The first and second terms in (2.7) describe the generation of supersaturation by adiabatic air cooling and supersaturation sink by condensation of water vapor on growing particles, respectively.

Near the cloud base, the value of the first term is much larger than that of the second one because of the lack of droplets, and the value of supersaturation in the cloud parcel rapidly increases with height. Nucleated droplets absorb water vapor decreasing supersaturation. As a result, supersaturation attains its local maximum at 10–30 m above the cloud base. In the point of supersaturation maximum \(dS/dt = 0\), and, consequently,
\( A_r / A_x \) \( W = dq_1 / dt \). The left side of the equality is known. Ghan et al. (1993, 1995), Bedos and Rosset (1996) and Abdul-Razzak et al. (1998) propose sequentially updated procedures to determine the minimum CCN size at the point of maximum supersaturation using the value \( dq_1 / dt \). In this way, the authors calculate the number of activated AP and the number of activated droplets. Thus, the droplet concentration is determined even without using (or knowing) the value of supersaturation maximum. The method seems to be very useful for precise determination of concentration of droplets nucleated at the cloud base.

Note, however, that droplet nucleation can take place above the cloud base as well. Such nucleation can be induced by an increase in supersaturation with height (see Section 3.5), and in case of lateral entrainment of the surrounding air and subsequent mixing with cloudy air (Section 3.4). These processes are important with regard to droplet spectrum broadening and precipitation formation and should be taken into account along with droplet nucleation at the cloud base. In addition, a vertical profile of supersaturation can contain two or more maxima (see, for instance, Fig. 4 in Roesner et al., 1990).

As it will be shown in Section 5, the initial droplet spectra have a profound effect on the cloud evolution. An adequate treatment of the formation of the spectrum is, therefore, of primary importance in cloud modeling.

*Technical notes.* (1) There is a problem related to utilization of parameterization (2.2) in numerical models. This expression is derived from laboratory measurements and other observations and does not contain time explicitly. In some models (e.g., Reisin et al., 1996), the number of CCN predicted by (2.2) is regarded as a maximum possible drop concentration, so that if drop concentration in a particular grid point exceeds the value predicted by (2.2), the process of new nucleation is not conducted.

Note that Eq. (2.2) (see ice nucleation parameterization below as another example) actually provides the distribution of CCN concentration with respect to supersaturation: \( dN / dS = kN_0 S^{k-1} \), where \( dN \) is the number of newly activated CCN when supersaturation in an air parcel increases by \( dS \). Let us assume that during a model time step \( dt \) an air parcel moves from the point \( \{ x-dx, y-dy, z-dz \} \) to the regular grid point \( \{ x, y, z \} \) with the velocity with components \( u, v, \) and \( w \) (Fig. 2). Let us assume that in the point \( \{ x-dx, y-dy, z-dz \} \) the concentration of AP was equal to \( N \) and supersaturation was equal to \( S(t-dt, x-dx, y-dy, z-dz) \). While the air parcel is moving, the value of supersaturation in it changes by \( dS \). The rate of CCN activation can be calculated as

\[
\text{d}N = \begin{cases} 
N_0 KS^{k-1} \text{d}S, & \text{if } \text{d}S > 0 \\
0, & \text{if } \text{d}S < 0
\end{cases} \quad (2.8)
\]

The value of \( dS \) can be written in the Eulerian coordinate frame as:

\[
\text{d}S = \left( \frac{\partial S}{\partial t} + u \frac{\partial S}{\partial x} + v \frac{\partial S}{\partial y} + w \frac{\partial S}{\partial z} \right) \text{d}t. \quad (2.8a)
\]

The number of droplets newly nucleated during a certain time step in a particular model grid point does not depend on the number of CCN activated in this grid point earlier (as it was in the approach used by e.g., Reisin et al., 1996), but depends on the
Fig. 2. Scheme of an air parcel track during one model time step from a point \( \{x-dx, z-dz\} \) to the grid point \( \{x, z\} \). During the motion of the parcel, supersaturation with it changes by \( dS \).

The rate of change in supersaturation in the air parcel reaching the grid point. The value \( dS \) in (2.8a) in a corresponding model grid point can be also calculated using a usual semi-Lagrangian numerical scheme. Thus, an adequate description of process of droplet nucleation should be based on (2.8) and (2.8a).

As shown above, (2.2) also provides the size distribution of CCN and nucleated droplets within a certain range of particle sizes. Thus, the size distribution of newly nucleated droplets should not be chosen in an arbitrary way.

(2) Special attention should be paid to the source of water for nucleated droplets. AP grow mainly below the cloud base at the expense of water vapor in the atmospheric boundary layer, and not at the expense of water vapor in the point where the droplet nucleation takes place. Nucleation is actually an instantaneous process, during which a wetted particle exceeds its critical size and becomes a droplet. The particle sizes can be already close to their critical size at the cloud base. An increase in the particle size during the act of nucleation can be negligibly small, as well as latent heat release by nucleation.

Contrary to it, when nucleation of droplets is assumed on CCN entering a cloud through its lateral boundary at high levels, or hygroscopic seeding of clouds by large particles is simulated, CCN grow at the expense of water vapor within a cloud layer.

2.1.2. Diffusion growth of liquid drops

2.1.2.1. Theory and observations. Diffusion growth of droplets and ice crystals is accompanied by the release of latent heat and is, therefore, the main energetic source of cloud updrafts. Droplets can grow by diffusion (condensation) to 10–20 μm in radius, depending on cloud type. Large droplets grow by diffusion very slowly, so that raindrop formation is due to drop collisions. The diffusion growth of droplets is described by Eq.
(2.1). For droplets with the radii exceeding a few microns, the last two terms are much smaller than the first one, and are usually omitted. As a result, in cloud models the continuous process of aerosol particles growth is artificially separated into two processes: droplet nucleation, when all terms in Eq. (2.1) are taken into account, but equilibrium \((dr/dt = 0)\) is assumed, and droplet diffusion growth, which is described by the simplified equation:

\[
\frac{dr}{dt} = \frac{1}{r} \frac{dS}{dF}.
\]  

(2.1a)

Due to this simplification, any supersaturations \(S > 0\) lead to droplet growth. A possible process of partial evaporation and "denucleation" of droplet under low values of \(S\) in areas of low or negative vertical velocity are excluded from consideration. Korolev (1995) attempted to analyze the role of the omitted terms on the drop spectrum evolution. This problem requires further investigation.

Eq. (2.1a) leads to the following two main specific features of the droplet diffusion growth.

(a) Eq. (2.1a) predicts a faster growth of the radii of smaller droplets than of larger droplets, and, hence, the narrowing of the initial drop size distribution with the distance above the cloud base (Mordy, 1959; Cooper et al., 1997). Typical values of supersaturation in cumulus clouds are supposed to range from 0.1% to 1% (e.g., Mazin and Shmeter, 1983). Under these conditions, the classical theory of condensation growth is unable to explain the formation of large cloud droplets (20–30-μm radii) within reasonable time. At the same time, large droplets of this size are regularly measured in situ observations (Brenguier, 1998). In fact, many in situ observations beginning with early measurements in the 1960s (Warner, 1969a, 1973) to the very recent ones (Brenguier, 1998) show broad mean droplet size distributions and the broadening of the drop size spectra with height. The droplet spectrum dispersion remains nearly constant with height and for typical cumulus clouds it is about 0.15–0.3 (Politovich, 1993). It should be stressed that the width of droplet spectrum is determined not only by the formation of the droplets with the radii exceeding the value that can be attained during adiabatic ascent (so called, adiabatic value), but mainly by the existence of droplets with the radii smaller than the adiabatic value (say, within the range from 1 to 10 μm), regularly observed (e.g., Warner, 1969a; Brenguier, 1998) in clouds at the distance of several hundred meters above the cloud base and leading often to the bimodal spectra formation. The existence of these small droplets (and bimodal spectra) in undiluted cores of cumulus clouds is as mysterious as the existence of the largest droplets.

(b) Droplets from a very narrow spectrum have similar fall velocities and hardly can collide. It means that the narrowing of the drop spectrum retards the coalescence growth of cloud droplets, regardless of the size reached as a result of the diffusion growth.

The problem of droplet spectrum broadening is discussed further in Section 3. Here we would only like to mention that there were many attempts to explain differential droplet spectrum growth by fluctuations of supersaturation in clouds using Eq. (2.3) (see review by Pinsky et al., 1999a). The main difficulty to explain the differential drop growth by diffusion is that the fluctuations of supersaturation caused by stochastic fluctuations of water vapor or by fluctuations of drop concentration related to the inertia
effects turn out to be short-lived, so that all droplets lifting within cloud updrafts experience nearly the same history of supersaturation fluctuations.

Shaw et al. (1998) assumed that the intermittent structure of turbulence in clouds leads to the formation of long-lived turbulent tubes, in which supersaturation differs significantly from that outside such turbulent vortices. The problem of applicability of this hypothesis to real clouds requires a better understanding of turbulence processes.

2.2. Drop growth by collection

2.2.1. Theory and observations

Drop collection growth is the main mechanism controlling rain formation in warm clouds. The process is described by the stochastic kinetic equation of collisions, which can be written as follows (Pruppacher and Klett, 1997):

\[
\frac{\partial f(m)}{\partial t} + \frac{\partial u f(m)}{\partial x} + \frac{\partial v f(m)}{\partial y} + \frac{\partial (w - V_t) f(m)}{\partial z} = 2 \int_0^{m/2} f(m - m') K(m, m - m') f(m') dm' - \int_0^{\infty} f(m) K(m, m') f(m') dm'
\]

(2.9)

where \( m \) is the drop mass, \( f(m) \) is the number distribution function \( f(m) dm \) is the number of drops per unit of volume within the range of masses from \( m \) to \( (m + dm) \), \( V_t \) is the terminal fall velocity of drops with mass \( m \). \( K(m, m') \) is the collection kernel which determines the probability of collisions of drops with mass \( m \) and \( m' \). The kernel can be written as:

\[
K(m, m') = \pi (r + r')^2 |V - V'| E_{\text{col}} E_{\text{coal}}
\]

(2.10)

where \( r \) and \( r' \) are radii and \( V \) and \( V' \) are the velocities of drops with the masses \( m \) and \( m' \), respectively. In a homogeneous flow, the difference between drop velocities is equal to the difference between terminal velocities. \( E_{\text{col}} \) is the collision efficiency, defined as the ratio of collision cross-section to the geometric cross-section \( \pi (r + r')^2 \). The coalescence efficiency, \( E_{\text{coal}} \), determines the fraction of collisions which results in drop coalescence.

First term on the right hand of (2.9) describes the rate of generation of drops with mass \( m \) by coalescence of drops with masses \( m' \) and \( (m-m') \). The second term describes the decrease of number of drops with mass \( m \) by coalescence with other drops.

In cloud models size distribution functions depend on coordinates and time, so that we added to Eq. (2.9) the corresponding terms determining the drop advection. Modelers used to include diffusion terms in (2.9) as well. The consequence of this on droplet spectrum formation will be discussed below.

As can be seen from Eq. (2.9) and (2.10), the collection rate is determined by the relative velocity between drops and by the collision and coalescence efficiencies, which are functions of relative velocity between drops and the drop sizes. In calm air, both values are determined by masses of colliding droplets.

Most studies dedicated to determination of the collision efficiencies have been conducted in 1970s using different numerical methods and laboratory measurements (Pruppacher and Klett, 1997). A characteristic feature of theoretical calculations is that
each study was concerned with a specific relatively narrow range of sizes of colliding drops. The determination of the collision efficiencies is a complicated hydrodynamic problem of drop interaction. Each drop induces a velocity field, which influences the motion of its counterpart. The velocity field induced by droplets with radii below 30 μm is well described by the analytical solution presented by Stokes (Kim and Karrila, 1991). Hocking (1959) calculated the force exerted between two droplets to determine collision efficiency. Davis and Sartor (1967) and Hocking and Jonas (1970) extended his approach to include rotation of the droplets. They assumed that inertial force is negligibly small in Stokes equations. Consequently, their approach can be only applied to situations when Reynolds number is close to zero (Re ≪ 1) (Lin and Lee, 1975). The effect of inertial force was taken into account by Klett and Davis (1973) using an analytical model based on modified Oseen’s equations made by Carrier (1953). They used the method for description of the velocity fields induced by drop collectors with the Reynolds number up to 5. However, this method is only valid for small droplets as well due to the inherited limitation of the Oseen’s equation (Lin and Lee, 1975). For larger drop-collectors the induced velocity fields are calculated numerically (e.g., Shafrir and Gal-Chen, 1971; Lin and Lee, 1975).

Because of difference in approaches and numerical schemes used, the values of the collision efficiencies obtained by different authors are often inconsistent (Pruppacher and Klett, 1997). The discrepancies are especially significant in the important case of small droplet collisions, as well as collisions of small droplets with larger ones.

Hall (1980) compiled results of different studies and tabulated them for use in microphysical models by means of interpolation. The values presented by Hall especially for small droplets seem greater than those reported by some other authors. Averaging of the values in the zone with extremely high gradients of the collision efficiencies (small ratios of sizes of collected and collecting drops) does not guarantee the obtaining of right values of the collision efficiencies. In addition, the table of Hall does not contain information of the collision efficiencies in drop pairs containing smallest drops.

Hence, it was desirable to recalculate the collision efficiencies within the frame of one approach for drops within as wide range of sizes possible. These calculations have been recently conducted by Pinsky et al. (1999b). The method is based on the matching of analytical solutions obtained by Stokes and Hamielec and Johnson, 1962 for different Reynolds numbers (drop sizes).

The method was used for calculation of the height dependency of the collision rate. Note that cumulus clouds can easily reach heights of several kilometers. The air density at 500-mb level is about half as large as at the surface (1000 mb). It is clear that drop terminal fall velocities increase with height because of a decrease in the air density and, consequently, a decrease in the drag force. It results in an increase in the difference in the drop terminal fall velocities and in an increase in the swept volume with height. Evaluations show that the increase in the collision kernel caused by the increase in the swept volume is about 10% to 25% depending on height and drop size. This effect is often neglected in cloud models.

The importance of another effect is not yet well recognized. This second effect consists in an increase in the collision efficiency with height. In fact, the collision
efficiency depends not only on the size of colliding drops, but also on the relative velocity between them. Mutual tracks of droplets about to collide depend on their moments. The increase of the momentum of one drop leads to deviation of the drop track from the track of airflow around the other drop, increasing the value of the collision efficiency. The fact that the collision efficiency is a function of air temperature and density was mentioned by Mazin and Shmeter (1983). However, the sensitivity of the collision efficiency to these parameters was underestimated in their book as well as in other studies. To our knowledge, there were no studies (except that of Pinsky et al., 1999b) dedicated to a detailed investigation of the sensitivity of the collision efficiency to increase in the interdrop relative velocity caused by a decrease in air density with height. Most laboratory measurements of the collision (more correctly, collection efficiency) have been conducted under pressures close to 1000 mb.

The detailed hydrodynamic calculations of the collision efficiencies under different heights conducted by Pinsky et al. (1999b) showed that for certain drop pairs the collision efficiency at 500 mb is twice as large as 1000 mb. The increase in the collision kernel (the rate of collisions) with height is caused, therefore, by a product of the increase in the swept volume and the collision efficiency. Fig. 3 shows the ratio of the

Fig. 3. The ratio of the collision kernels at 500- and 1000-mb levels as seen from results of calculations by Pinsky et al. (1999b). One can see that the increase of the collision kernel with height is rather significant and different for different drop pairs. It is the strongest for droplets 5 to 10 μm colliding with comparably small drop collectors of 15 to 30 μm in radius.
collision kernels at 500- and 1000-mb levels as calculated by Pinsky et al. (1999b). One can see that the increase of the collision kernel with height is rather significant. It is the strongest for droplets 5 to 10 μm colliding with comparably small drop collectors of 15 to 30 μm in radius. For these drop pairs, the collision kernel at 500-mb level is more than twice as large as that at 1000-mb level. The results of the spectrum evolution simulations at different heights indicate significant acceleration of the collision process with height (Fig. 4). The acceleration is so significant that it should be taken into account in cloud models, when the droplet growth by collision is considered.

Note that laboratory measurements determine actually the collection efficiency, which is product of the collision and coalescence (sticking) efficiencies. Beard and Ochs (1984, 1993) showed that the coalescence efficiency decreases with increasing drop sizes from 100% for drop-collector radius \( R < 25 \mu m \) and the collected droplet radii \( r < 10 \) to less than 50% for \( R > 800 \mu m \) and \( r > 40 \mu m \). There is some laboratory evidence that the coalescence efficiency increases with the air humidity and equal to 1 at 100% relative humidity (Borovikov et al., 1961) and with height (Beard et al., 1998). The combined effects of the increase of the collision and coalescence efficiencies with height on the collision rate have not yet been investigated.

2.2.1.1. Technical notes. (1) The following are the three schemes widely used in solving the stochastic kinetic equation of collisions.


(b) The Berry and Reinhardt (1974) method used, for instance, by Kogan (1991), Khain and Sednev (1996), and Alheit et al. (1990);

(c) The method of moments (Tzivion et al., 1987) is used, for instance, by Reisin et al. (1996) and Yin et al. (2000).

The Kovetz and Olund (1969) method induces large numerical broadening leading to too fast formation of raindrops (see discussion in Cooper et al., 1997), so that its use is not recommended.

The method by Berry and Reinhardt (1974) is rather precise when a drop spectrum evolution is simulated. The disadvantage of the method consists in the necessity to calculate size distribution functions in points not coinciding with the regular points of the model mass grid. These values are calculated using a six point Lagrange interpolation formula. Sometimes, when a mixed phase cloud is simulated, size spectra of ice particles become narrow or multi-modal. In this case, the precision of the scheme decreases substantially and some loose of mass (up to 30%) and significant errors in the reproduction of size distribution functions are possible.

Method of moments is more accurate (Tzivion et al., 1999), but it requires solving equations for both masses and concentrations of each bin, while the Berry and Reinhardt (1974) method is based on solving the equations only for bin masses.

A new computationally effective scheme of solving stochastic collision equations has been suggested recently by Bott (1998). We tested the approach carefully against the precise benchmark method (Seesselberg et al., 1996). The results of the comparison showed an excellent agreement in all testing experiments. The scheme suggested by Bott conserves mass, appeared to be very stable working under different initial droplets spectra and very effective from a computational point of view, permitting one to use...
Fig. 4. (a,b) Evolution of droplet size spectrum at (a) 1000 and (b) 500 mb. One can see significant acceleration of the collision process with height. $G$ is the mass density distribution function determined as in Berry and Reinhard (1974).
comparably large time steps (10 to 20 s). The scheme revealed many advantages in case of drop–ice and ice–ice collisions (Khain et al., 1999c). We would like to recommend the use of this method in cloud models.

(d) In numerical models, the collision kernels of particles belonging to the same mass bin are usually assumed equal to zero. Note, however, that in case of a comparably low resolution of mass grids, each grid point (each bin) is responsible for a range of drop sizes. For instance, a 30-μm radii drop can be “responsible” for drops with radii ranging from, say, 25 to 35 μm. Therefore, collisions of drops belonging to this mass bin actually describe collisions of drops ranging from 25 to 35 μm, the collision kernel of which is not equal to zero. It means that the values of the collision kernel for drops belonging to the same bin has to be determined by averaging the collision kernels of drops within this mass bin, but not set equal to zero.

3. Warm rain microphysics: spectral broadening

3.1. Background notes

We review this problem in a separate section because (1) the problem is of significant importance of the problem in cloud microphysics and (2) it is closely related to both diffusion and collision drop growths.

As was mentioned above, the classical equation of diffusion growth predicts narrowing of the droplet size spectra. Besides, the theoretical evaluations (Jonas, 1996) show that in a typical cumulus cloud with a liquid water content of 1 g kg⁻¹ the largest drop in drop size spectrum reaches a radius of 20 μm in 1 h. It is known, however, that such clouds precipitate in only about 20–25 min after their formation.

Note that the problem of the droplet spectrum broadening contains two problems: the formation of droplets with size exceeding theoretical limit that droplets can attain during adiabatic ascent by diffusion, and with sizes below the adiabatic value at distance as large as a few hundred meters above cloud base, often leading to the formation of bimodal droplet size distributions all over the cumulus clouds including undiluted cloud cores (e.g., Squires, 1958; Warner, 1969a,b; Brenguier, 1998).

Many investigators looked for the cause of droplet spectrum broadening in stochastic fluctuations of humidity (e.g., Belyaev, 1961; Levin and Sedunov, 1968; Voloshuk and Sedunov, 1977).

We illustrate difficulties arising from this assumption using as an example a case of monodisperse droplet spectrum at the cloud base. Supersaturation in clouds rapidly attains its quasi-steady value, determined as (e.g., Cooper, 1989):

\[ S_{\text{qs}} = \frac{C W}{r N}, \]  

(3.1)

where \( W \) is the vertical velocity, \( N \) is the droplet concentration, \( C \) is the coefficient depending slightly on temperature. From (2.7) and (3.1) we have:

\[ r^2 \frac{d r}{d t} = C_i W \]  

(3.2)
where $C_1$ is a coefficient that is slightly dependent on temperature and air properties (assuming invariable drop concentration). Let us assume that an air parcel rises from the cloud base ($z = 0$) to a height $h$ above the cloud base. Integrating Eq. (3.2) with respect to time yields:

$$m(z = h) - m(z = 0) = C_2 h$$

(3.3)

where $m$ is the droplet mass, $C_2$ is a coefficient slightly depending on height. Expression (3.3) shows that an increase in the drop mass depends on the distance above the cloud base only. It means that a droplet spectrum, being monodisperse at the cloud base, remains to be monodisperse at any height level independently of both supersaturation fluctuations (caused by vertical velocity fluctuations) and on drop tracks. Mazin and Smirnov (1969) showed that the shape of a multidisperse droplet spectrum depended only on the height above the cloud base and did not depend either on the tracks of different droplets or on the time that droplets needed to reach the level above the cloud base. This result is a direct consequence of close relationship (3.1) between vertical velocity and supersaturation in clouds. Thus, any theory claiming to explain the broadening of droplet spectrum must break down this proportionality.

In other words, it seems that only mechanisms influencing the values of supersaturation through fluctuations of droplets concentration can lead to droplet spectrum broadening.

### 3.2. Mixing with the surroundings and fresh nucleation

Mixing with the surroundings is usually thought to be one of most likely mechanisms for spectrum broadening. Two main mechanisms of mixing were suggested: lateral homogeneous mixing (Mason and Chien, 1962; Warner, 1973; Mason and Jonas, 1974; Lee and Pruppacher, 1977; Lee et al., 1980), and inhomogeneous mixing (Baker et al., 1980; Baker and Latham, 1982; Jensen and Baker, 1989; Baker, 1992). According to the concept of lateral homogeneous mixing entrained air spreads laterally at an infinite speed through the cloud. According to the concept of inhomogeneous mixing, parcels ascend adiabatically from cloud base, until at some level where they mix with the clear air environment. The entrainment process is discrete and it occurs on scales of one to several hundred meters. Localized mixing leads to a decrease of droplet sizes and to an increase of the number of the smallest drops in the droplet spectrum within particular cloud volumes. Then, turbulent mixing supposedly leads to the homogenization and formation of a region of uniform characteristics, but, with droplet spectrum, which is broader than before. It is also assumed that inhomogeneous mixing can lead to local increase in supersaturation and to the formation of largest droplets.

Formation of the smallest droplets is often attributed to nucleation of droplets during entrainment of drop-free air into a cloud through lateral boundaries (Roesner et al., 1990; Brenguier and Grabowski, 1993; Su et al., 1996, 1998; Kruger et al., 1997; Pinsky et al., 1999a).

Kruger et al. (1997) and Su et al. (1998) developed a model in which four processes are parameterized: parcel ascent, entrainment of air from lateral boundary, turbulent mixing, and droplet growth. Entrainment is assumed to occur in discrete events. During
each event, a randomly selected region of the parcel is replaced with environmental air. Entrained blobs were assumed to have sizes from 2 to 200 m in different versions of their model. The model generates a band-type droplet spectra in case of instantaneous mixing (each entrainment event, being accompanied by droplet nucleation, leads to the formation of a new isolated peak in the droplet size spectra), and a wide plateau-like spectra with the droplet size from about 2–18 μm and to 23–25 μm at 1100 and 1500 m above the cloud base, respectively, in case of finite-rate mixing (Su et al., 1998).

There is no consensus as to the role of lateral mixing (either homogeneous or inhomogeneous) in reaching the observed droplet size distributions (see, e.g., discussions of Manton and Warner (1982) and Jonas and Mason (1982); and more recently, Hudson and Yum (1999) and Telford (1999)).

Note that because of many uncertainties of the complicated process of entrainment and subsequent turbulent mixing, numerical models based on the entrainment/mixing mechanism contain many assumptions and simplifications. As a result, the shape of simulated spectra turns out to be crucially dependent on such poorly known parameters as: the size of entrained blobs, the frequency of the entrained events, the height levels at which the entrainment takes place, the propagation speed of turbulent blobs into a cloud, etc. Many details of the turbulent mixing process accompanied by drop nucleation and evaporation remain to be unknown or poorly understood. A proper description of a very complicated process of mixing requires a better understanding of the structure of cloud turbulence on small spatial and time scales, deep insight in motion of inertial particles within a turbulent flow, as well as detailed description of processes of nucleation/de-nucleation. More investigations in this direction are required.

### 3.3. Multi-parcel mechanism of the droplet spectrum formation

In many studies (e.g., Telford and Chai, 1980, 1983; Telford and Wagner, 1981; Telford et al., 1984; Ferrier and Houze, 1989) it is stressed that entrainment is greatest at cloud top, in disagreement with widely used entrainment parametrizations through the lateral boundaries. It was concluded that single parcel models, in which evolution of droplet spectra is determined by nucleation of CCN entering through lateral cloud boundaries, are unable to simulate the evolution realistically.

Mason and Jonas (1974) and Roesner et al. (1990) assumed that a realistic droplet spectrum can form only in series of subsequent parcels, in which a new parcel ascends within the surroundings formed by earlier parcels. Taking into account many uncertainties in the theory of inhomogeneous mixing, Roesner et al. (1990) use a parcel model with isotropic and instantaneous mixing with variable surrounding conditions caused by previous parcels. When a set of ascending parcels is considered within the frame of a Langrangian parcel model, a complicated problem of their interaction arises, because it is not quite clear what properties inside the air (temperature, vertical velocity, droplet and CCN size distributions, etc.) are “left behind” by the previous parcel. Roesner et al. (1990) keep the values within a previous parcel “frozen” at each level and use them to represent a new environment between the condensation level and the maximum height that the parcel has reached.
In the simulations conducted by Roesner et al. (1990), bimodal droplet spectra are caused by the entrainment of fresh CCN in the vicinity of the top of the last parcel of the parcel set, only after when the parcel had emerged from the air of the previous parcel. At lower levels, entrainment of residual air containing drops and interstitial AP does not lead to new nucleation in the parcel.

Thus, these studies consider the mechanism of lateral entrainment of dry air with fresh nuclei as the main mechanism of droplet spectrum broadening.

Note in this connection the following points.

(a) Bimodal spectra were observed in undiluted cloud cores (Warner, 1969a), where effects of lateral mixing are hardly effective.

(b) The concentration of CCN, especially larger ones, decreases significantly with height. Many authors (e.g., Mazin et al., 1989) report an exponential decrease of CCN concentration with height with characteristic scale of 1.5 km. Warner (1969a) found that bimodal spectra formed in maritime clouds at 2 km above cloud base. Alkezweeny (1995) observed the formation of bimodal spectra at the heights of several kilometers above the cloud base. It is not very clear why these bimodal spectra do not form at lower levels, where the process of mixing and entrainment must be effective and CCN concentration in the environment is higher.

(c) The mechanism of lateral mixing is hardly responsible for the strong reduction and even depletion of the fraction of samples having a bimodal distribution with an increase in the cloud layer stability observed by Warner (1969a).

This analysis indicates that lateral mixing is not an exhaustive process (and, possibly, not a dominating one) of droplet spectra broadening and bimodal spectra formation.

3.4. Fluctuations of drop concentration caused by updraft velocity fluctuations at cloud base

Several investigators (e.g., Kabanov et al., 1970; Warner, 1969b; Cooper, 1989; Korolev and Mazin, 1993; Korolev, 1994) pointed out that fluctuations of drop concentration can be related to the fluctuations of vertical velocity at the cloud base. Different vertical velocities at the cloud base, as well as fluctuations of CCN concentration, temperature and humidity can lead to different values of the supersaturation maximum and different droplet concentrations in rising updrafts. According to this mechanism, further mixing of unimodal spectra leads to the formation of wider bimodal (or multi-modal) spectra.

3.5. In-cloud activation of cloud condensation nuclei ascending from the cloud base

It is well known that updraft speed at the cloud base determines the maximum supersaturation and droplet concentration in the vicinity of the cloud base (e.g., Warner, 1969a; Rogers and Yau, 1989). Note that the maximum in supersaturation leads to the activation of only a certain fraction of AP, so that ascending cloud parcels contain both nucleated water droplets and non-activated AP at any level above the cloud base.

In Section 2.1.1, we saw that according to (2.7) supersaturation attains its local maximum at about 10 m above the cloud base. Nucleation of CCN ascending from the
cloud base is possible therefore if supersaturation exceeds the cloud base maximum at some height. Nucleation of new droplets distances as large as a few hundred meters above the cloud base would lead to (a) the droplet spectra broadening through the formation of smaller droplets and (b) the formation of bimodal droplet spectra without any mixing with other cloud parcels or cloud surroundings.

Thus, the problem of in-cloud nucleation of new droplets on APs entering cloud through cloud base reduces to the following two questions: (1) can the supersaturation within a rising cloud parcel exceed the maximum it reaches at cloud base? And (2) is the reservoir of potential CCN large enough to allow new droplet nucleation in case the supersaturation within a rising cloud parcel exceeds the maximum near the cloud base?

Khain and Pinsky (2000a,b) show that for a wide range of conditions, the answers to both questions are positive, i.e., the supersaturation within a rising cloud parcel can exceed the maximum it reaches at cloud base, and the reservoir of potential CCN is usually large enough to allow new droplet nucleation.

The mechanism proposed is simple enough and easily can be reproduced by most cloud models. Thus, a question arises, why was this mechanism not found earlier? We suppose three main reasons for this situation.

3.5.1. Utilization of a preset profile of the vertical velocity

In most studies dedicated to the investigation of the cloud droplet spectrum formation, the vertical velocity of an ascending cloud parcel is assumed to be height independent (Warner, 1969a; Rogers and Yau, 1989). As is clear from (2.7), the second term increases with height, so that the assumption that $W$ is height independent leads to a decrease in supersaturation with height. As a result, no new activation of CCN ascending within a cloud parcel takes place (in case of no lateral mixing with surroundings), and the formed spectra are narrow in accordance with the classical theory.

It is also clear that supersaturation can increase with height if the first term on the right side in (2.7) grows faster that the second one, i.e., if the velocity acceleration exceeds a certain value. In some experiments Warner (1973) used the profile of vertical velocity given as $W = W_0 + Cz^{1/2}$ corresponding to the assumption that the parcel buoyancy $B$ does not depend on height. This acceleration turns out to be insufficient to allow supersaturation to exceed the cloud base maximum. Again, as in case $W =$ const, the activation of CCN ascending from the cloud base did not take place.

Note that the assumption $B =$ const corresponds to a height independent difference between the cloud parcel temperature and its surroundings (the loading is negligible at small distances above the cloud base of a developing cloud). It is well known, however, that the difference is minimum at the cloud base and increases with height within a few kilometers above the cloud base. Thus, updraft cloud parcel velocity within this layer grows with height faster than $z^{1/2}$.

3.5.2. Utilization of large values of vertical velocity at cloud base

It is known (e.g., Fig. 7.4 in Rogers and Yau, 1989) that the supersaturation maximum near cloud base increases with the vertical velocity at cloud base $W_0$. The probability for supersaturation to exceed this maximum during the parcel ascend
decreases, therefore, with an increase in $W_0$. According to results of Khain and Pinsky (2000a,b), the values $W_0 = 1 \text{ m s}^{-1}$ can be considered as a typical value, separating the regimes with and without in-cloud nucleation in not very deep cumulus clouds.

In many studies, the values of vertical velocities at cloud base were set too large to allow in-cloud supersaturation to exceed the cloud base maximum. For instance, Roesner et al. (1990) set $W_0 = 1 \text{ m s}^{-1}$ for simulation of low intensity clouds with intense lateral homogeneous mixing. Cooper et al. (1997) used $W_0 = 3 \text{ m s}^{-1}$. Under the thermodynamic conditions used in these studies, the droplet spectra could be broadened, only allowing the lateral mixing and related droplet nucleation.

The mean (or maximum) cloud base vertical velocity can, of course, exceed 1 m s$^{-1}$ in cumulus clouds. Note, however, that vertical velocities within and at the base of cumulus clouds experience strong spatial and time variations (e.g., Warner, 1969a, 1970, Warner, 1973; Cooper, 1989; Mazin et al., 1989). Vertical updraft velocities usually tend to decrease to periphery of cloud updrafts, often replacing by downdrafts. Besides, one can expect smaller velocities at the earlier stage of cloud formation. Thus, along parcels with high values of cloud base vertical velocities, there exists a significant number of parcels with much lower velocity at the cloud base.

### 3.5.3. Lack of the reservoir of small aerosol particles

An increase in supersaturation in excess of the cloud base maximum was found earlier by Lee and Pruppacher (1977) using a parcel model with and without dry air entrainment. However, they did not find any in-cloud nucleation discussed above. We attribute this result to the fact that they used CCN with sizes exceeding 0.2 μm (see Fig. 1 in that paper). There were no smaller particles in the CCN spectrum used. At the same time Lee and Pruppacher used a significant vertical velocity at the cloud base (1 m s$^{-1}$). Under this condition the supersaturation maximum near the cloud base reached 0.3%, so that all CCN in the CCN spectrum were activated. Indeed, the supersaturation needed for activation of 0.2-μm radius CCN is only about 0.01% (Rogers and Yau, 1989, p. 89). As a result, the reservoir of potential CCN was totally exhausted near cloud base. Note, however, that according to Pruppacher and Klett (1997, p. 289) the spectra of aerosol particles (both continental and maritime ones) usually contain small CCN, so that nucleation of new droplets continuously takes place with an increase in supersaturation up to several percents.

Khain and Pinsky (2000a,b) investigated the droplet spectra broadening and bimodal spectra formation by nucleation of CCN ascending from the cloud base using a precise microphysical model of a cloud parcel.

The equation system includes the equation for diffusion growth (2.1), as well as equation for supersaturation (2.7), temperature and updraft speed of a rising cloud parcel (Pruppacher and Klett, 1997). To describe the size distribution of non-activated AP and droplets, 2000 mass bins were used, with the mass increment increasing initially exponentially up to the maximum mass, corresponding to a droplet radius of 20 μm. The aerosol size distribution is taken in the form suggested by Respondek et al. (1995).

Fig. 5a,b shows the vertical profiles of updraft speed and supersaturation, respectively, for cloud parcels with different values of updraft speeds at the cloud base $W_0$ ($z=0$). We suppose the profiles $W(z)$ obtained are typical of undiluted updrafts in
strong cumulus clouds. In Fig. 5b the vertical profiles of supersaturation calculated under the assumption of constant (height-independent) updraft speeds are presented for comparison. The analysis of Fig. 5b shows that: (a) for $W = \text{const}$ supersaturation decreases monotonically with height above the level in the vicinity of the cloud base where supersaturation attains its maximum in accordance with the classical results; and
(b) if a realistic increase in vertical velocity with height above the cloud base is taken into account, supersaturation in the cloud updraft can exceed the local maximum at the cloud base.

It is clear that within the layer, where supersaturation increases with height, remaining higher than the local maximum at the cloud base (shaded zone in Fig. 5b), new

![Graph](image)

**Fig. 6.** (a) The cloud spectrum formed in the cloud parcel having low speed at the cloud base. (b) The same as in (a), but for high vertical velocity at the cloud base.
in-cloud nucleation and the formation of a bimodal droplet size spectrum take place, which consequently leads to droplet spectrum broadening.

Droplet concentration and the shape of droplet size spectrum depend, therefore, on the vertical velocity at the cloud base $W_0$ and on the acceleration of updraft. For instance, for high updrafts at the cloud base all droplet nucleation is concentrated in the vicinity of the cloud base with no new nucleation above. Droplet spectrum in this case reveals the features found in previous studies: the droplet size spectrum is narrow and becomes narrower with height (no in-cloud nucleation) (Fig. 6b). The cloud spectrum formed in the cloud parcel having low speed at the cloud base (Fig. 6a) differs crucially from that shown in Fig. 6b. One can see that in-cloud nucleation leads to the formation of bimodal spectra. Droplets nucleated near the cloud base form the right maximum in the spectra. Newly nucleated droplets within the cloud form the left maximum.

Fig. 7 shows the vertical profile of the dispersion of the drop size spectrum (the ratio of the drop spectrum width to the mean drop radius) averaged over about 500 cloud parcels arising from the cloud base with different velocities. We can see that in-cloud nucleation being taken into account allows us to explain the formation of size spectra with dispersion of 0.15–0.25, regularly observed in cumulus clouds. Thus, the values of droplet spectrum width, as well as the droplet size spectrum dispersion of simulated spectra, turn out to be realistic and typical of in-situ observed spectra. Note that the increase in the droplet spectrum dispersion is related to the formation of the smallest droplets by nucleation. This process is unable to explain the formation of large superadiabatic droplets.

In-cloud nucleation and the formation of bimodal spectra should be observed more often in cases where updraft speed at the cloud base is low (the cloud base supersatura-

![Fig. 7. The vertical profile of the dispersion of the drop size spectrum averaged over about 500 cloud parcels arising from the cloud base with different velocities.](image-url)
The discussion above shows that the existing hypotheses claiming to explain formation of the largest droplets by diffusion are subject to contradictions. We now turn to the hypotheses that include drop coalescence as an important factor of droplet spectrum formation at the early stage of cloud development. Two processes will be analyzed:

(a) collisions of large droplets arising on giant and ultragiant nuclei (GN, UGN) and
(b) self-broadening of droplet spectrum by collisions of cloud droplets.

### 3.6. Giant and ultra-giant nuclei

Possible influence of giant nuclei on the warm rain initiation was analyzed in many studies (e.g., Beard and Ochs, 1993; Lasher-Trapp et al., 1998; Yin et al., 2000) that tried to determine under what conditions drops arising on giant (GN) and ultra-giant nuclei (UGN) with radii larger than 15–20 μm can collect cloud droplets and grow to raindrop sizes. If this mechanism were effective, the process of rain formation would become linear and could be described by the equation of continuous growth (as it is done, for instance, by Lasher-Trapp et al., 1998). Note that the diffusion growth of droplets with radii above 20 μm is very low, so that their initial size at cloud base should be actually the same as at large distances above the cloud base.

Under the assumption that the formation of raindrops takes place on giant and ultragiant nuclei, the concentration of such nuclei must be of the order of that of raindrops (100–1000 m⁻³). The first problem therefore, is to find such GN and UGN in required concentrations at cloud base, and to establish how often such concentrations occur at the base of clouds at the earlier stage of cloud development. The concentration and size of UGN depend on the type of underlying surface (ocean versus soil) and on the low-level wind speed (Woodcock, 1953; Blyth et al., 1998). For example, during the Small Cumulus Microphysics Study (Florida) particles of radii up to 40 μm were observed in clear air when the low-level wind was blowing from the Atlantic Ocean. However, when the wind was blowing from the Florida mainland the largest particles were smaller than 15–20 μm in radius (Blyth et al., 1998). The size of the particles depends on humidity in the sub-cloud air. If the sub-cloud layer is dry, giant salt particles do not serve as CCNs because their growth by condensation is low (Beard and Ochs, 1993). The concentration of UGN generally decreases with height. There are, however, a few signs of their presence around clouds. For example, Laird et al. (1998) revealed from the data obtained during the Small Cumulus Microphysics Study (Florida) concentration of large particles of 25 and 50 μm in radius with concentrations of 76 and 2 m⁻³ respectively. Note that it is still a challenge to demonstrate experimentally the presence of UGN during the initial stage of cloud development: existing particle
counters are not well suited because their sampling section is far too small for measuring very low concentrations with a significant confidence level (Brenguier and Chaumat, 1999).

Results of numerical simulations of the effects of GN and UGNs on rain initiation are rather contradictory (Mazin and Shmeter, 1983; Johnson, 1993; Jonas, 1996; Blyth et al., 1998; Ochs et al., 1998; Lasher-Trapp et al., 1998). These discrepancies can be attributed to differences in UGN spectra used, as well as to difference in cloud water contents (CWC) and the sizes of cloud droplets.

We illustrate the evolution of the initial droplet spectrum by solving the stochastic equation of collisions using the approach developed by Bott (1998). We use 400 mass (radii) bins to resolve droplet size distribution from 1 to 2000 µm. Such a high resolution has not been used earlier. In Fig. 8 the evolution of the initial droplet spectrum centered at radius of 7 µm and CWC = 1 g m⁻³ (concentration of droplets is 1100 cm⁻³) is shown in cases of (a) no large droplets in the initial droplet spectrum and (b) when the additional distribution of large droplets was added to the initial spectrum of small droplets. The mass distribution of these large droplets was centered at radius of 20 µm. Concentration of droplets with radii above 20 µm was set at 400 m⁻³. One can see that even under such unrealistically large concentration of large droplets the rain production is comparably slow: about 8% of the whole LWC transferred to raindrops with radii of 500 µm in 40 min. Thus, GN and UGN hardly can be responsible for raindrop formation in case of when most cloud droplets are comparably small and CWC is not very large. This result agrees well with earlier evaluations conducted by Mazin and Shmeter (1983), Johnson (1993) and Jonas (1996), who stressed very low rate of collision growth of droplets with radii 20–30 µm under CWC of 1–2 g m⁻³.

It is clear that an increase in the droplet size along with the value CWC (say, up to 5 g m⁻³ as in Lasher-Trapp et al., 1998) must lead to an increase in the contribution of GN and UGN to rain formation. In case of very narrow droplet spectra and high CWC, the effect of the GN and UGN would be dominating (Cooper et al., 1997), because of the lack of interaction between small droplets of similar size.

Here, however, another problem arises: in many clouds, the width of drop spectrum is nearly proportional to the mean drop radius. For example, according to Warner (1969a) and Politovich (1993), the drop spectrum dispersion remains nearly constant with height with typical value in different cumulus clouds of 0.15 to 0.3. It means that droplet size distribution centered at, say 12-µm radius contains significant number of drops with radii exceeding 15–20 µm. In case of high CWC, the concentration of such droplets exceeds the possible concentration of drops that could arise on GN and UGN, and interaction between cloud droplets formed during cloud development begins to be dominating. As an example, Fig. 9a,b presents the evolution of initial wide droplet spectrum centered at 12 µm. The CWC was set equal to 1 g kg⁻¹ as in case of Fig. 8. Fig. 9a shows the spectrum development in case of neither GN nor UGN were assumed. Fig. 9b shows the drop evolution, when the concentration of drops with radii above 20 µm was set as large as 400 m⁻³. The comparison of the rates of drop spectra evolution in these two cases shows that in case of comparably wide droplet spectrum the process of spectrum self-broadening is dominating, so that the additional amount of UGN does not contribute significantly to the acceleration of the spectrum development. This last
Fig. 8. (a,b) Evolution of the droplet size spectrum initially centered at 7 \( \mu \)m, when (a) no large droplets are included and (b) a distribution of large droplets was added to the initial size spectrum of small droplets. Line type are as defined in Fig. 4.
Fig. 9. (a,b) Evolution of the size droplet spectrum initially centered at 12 μm. (a) Neither GN nor UGN are assumed. (b) A size distribution of large droplets with concentration of droplets exceeding 30 μm in radius of 400 m$^{-3}$ is added to the size spectrum of small droplets.

Example illustrates the formation of raindrops in a maritime cloud, in which the wide spectrum develops by cloud droplet collisions and GN do not play a significant role. This conclusion agrees with results obtained by Yin et al. (2000).
Thus, although the role of GN and UGN can be important in some cases, it is unlikely to be the universal dominant mechanism of drop spectrum broadening and raindrop formation.

Note in addition that drop spectrum broadening is usually understood as the formation of a comparably large concentration of droplets with radii 15 to 20 $\mu$m (Brenguier, 1998). In case when initial radii of drops forming on GN and UGN exceeds 20 $\mu$m, the process of their collisions with smaller droplets cannot be referred to as to droplet spectrum broadening in its usual meaning.

3.6.2. Small droplet collisions in a turbulent flow

A potentially important mechanism that can lead to the formation of larger droplets is the process of collisions between cloud droplets. Theoretical calculations and laboratory measurements (Pruppacher and Klett, 1997) showed, however, that the collision efficiencies of small droplets (with radii less than about 20 $\mu$m) in calm air are too small to provide any drop spectrum broadening within a reasonable time. Note that all calculations and measurements, discussed in Section 2, have been conducted for calm air conditions.

Recent laboratory experiments in wind tunnel conducted by Vohl et al. (1996, 2000) showed a significant acceleration of drop growth rate by collisions in a turbulent flow. Although these laboratory measurements were related to a few micrometer-radii droplets collected by small raindrops (exceeding 60 $\mu$m in radius), the results provided the reliable evidence of turbulence effects.

The turbulence effects on the motion and interaction of inertial particles were theoretically studied by Khain and Pinsky (1995, 1997), Pinsky and Khain (1996, 1997a,b), Pinsky et al. (1999c, 2000), and Zhou et al. (1997, 1998). Inertial droplets moving within a turbulent flow deviate from air streamlines. Differential drop response to the flow shears and accelerations leads to the formation of droplet–droplet relative velocities. The magnitude of the inertia induced relative velocities is determined by droplet mass and turbulence intensity. Pinsky et al. (1999c) calculated the collision efficiencies of small droplets in both calm air and turbulent flow and found that in the latter case, the collision efficiencies were significantly higher. Fig. 10 shows the mean values of the collision efficiencies of droplets smaller than 10 $\mu$m in radii. To facilitate comparison, the collision efficiencies for pure gravity case are presented as well. The calculations were conducted for the intensity of turbulence typical of early cumulus clouds (the dissipation rate is 100 cm$^2$ s$^{-3}$). One can see that the collision efficiencies in a turbulent cloud flow can be significantly (up to 10 times, in our example) larger than in the pure gravity case.

To illustrate the effects of turbulence on the droplet size spectrum evolution, the stochastic collision equation was solved using the Bott (1998) scheme. Fig. 11 shows the spectra formed at 12 min in a turbulent flow (curve B) and in a calm air (curve A). While no spectrum broadening was observed in the pure gravity case, droplets as large as 40 $\mu$m in radii formed in the turbulent flow toward 12 min. Further calculations of the spectrum evolution showed the formation of raindrops in the turbulent flow toward 30 min, and no raindrop formation in a calm air.
Fig. 10. Mean values of the collision efficiencies of droplets smaller than 10 μm in radii and 10 μm drop-collector in (A) a turbulent flow (the dissipation rate is 100 cm$^3$ s$^{-1}$) and (B) for pure gravity case (Pinsky et al., 1999c). $r$ and $R$ are the radii of collected droplet, and drop-collector, respectively.

The relative contribution of GN and UGN and collisions between small cloud droplets to rain formation depends on the width of the cloud droplet spectrum. That is why, the precise determination of the width is so important for the correct assessment of microphysical processes in clouds.

Note that coalescence of cloud droplets can explain the formation of larger droplets in the droplet spectra (exceeding adiabatic values), but is unable to explain the existence of comparably high concentration of the smallest droplets of 1 to 7 μm in the droplet spectra within undiluted cloud cores.

3.6.3. Fine cloud structure and droplet concentration inhomogeneity

Pinsky and Khain (1996) investigated drop motion in a turbulent flow using a stationary model of isotropic turbulence. The existence of areas of two types within a
turbulent flow was revealed. Due to their inertia, drops tended to leave the areas of enhanced flow curvature to concentrate in the areas of low curvature. Spatial scales of drop concentration inhomogeneities were found to be dependent on drop mass (inertia). For raindrops of a few hundred of microns in radius, the characteristic scales are of several meters and even tens of meters. For small cloud droplets, these scales are of one to a few centimeters.

Earlier, Maxey (1987) and Wang and Maxey (1993) demonstrated the formation of concentration inhomogeneity of inertial particles moving within turbulent flows. In the last study, the direct numerical simulation has been used for turbulent flow generation. Pinsky and Khain (1997b) and Pinsky et al. (1999a) theoretically showed that inhomogeneities of drop concentration were most pronounced for 100-μm radii drops.
Fluctuations of 10- to 15-μm radii droplets were found intense enough as well, attaining 20% to 40% of the mean drop concentration. The characteristic spatial scale of these fluctuations of droplet concentration is equal to one or a few centimeters; hence, clouds have fine structure ("inch clouds").

Note that the problem of the existence of small-scale concentration fluctuations in clouds (so called, preferential concentration) remains a controversial issue in the cloud-physics community (see a comment by Grabowski and Vaillancourt, 1999 on the paper by Shaw et al., 1998). Recently, these centimeter-scale drop concentration fluctuations have been retrieved from a series of drop-arrival times providing in-situ measurements using the Fast FSSP (Pinsky et al., 1998a; Pinsky and Khain, 2000). The RMS amplitude of small-concentration fluctuation in a cumulus cloud analyzed turned out to be 30% of the mean droplet concentration, which agreed well with the theoretical evaluations of Pinsky et al. (1999a).

The mechanisms of formation of concentration inhomogeneity of inertial particles in different turbulent flows were studied recently by Elperin et al. (1996), Zhou et al. (1997) and Wang et al. (2000).

The process of collisions is known to be more effective in case of inhomogeneous spatial distribution of particles (e.g., Voloshuk and Sedunov, 1975; Kasper, 1984). Zhou et al. (1997) demonstrated a significant increase of the collision rate of inertial particles using a turbulence model based on the solution of the Navier–Stokes equations.

As follows from the stochastic kinetic equation of collisions, the frequency of drop collisions is proportional to the product of drop concentration. Thus, the frequency of collisions increases in the areas of enhanced droplet concentration by a factor of 1.7 to 2.

Pinsky and Khain (1997b) assumed the existence of a “self-concentration” process of droplet collisions: larger cloud droplets being formed tend to concentrate in certain volumes, where their concentration turns out to be much higher than the averaged concentration of such drops. The increase in the concentration of these droplets increases the probability of their collisions with the formation even larger droplets. These droplets, in their turn, tend to concentrate with even higher rate, etc. The main idea of the assumption is that the fast droplet spectrum broadening with formation of rain drops takes place in the regions, where the concentration of larger clouds is higher than the mean concentration of such drops. This process was simulated by Pinsky and Khain (1997b) using a simple model. A faster broadening of the “mean” spectrum was obtained, as compared to that of homogeneous droplet concentration. Further studies are required to investigate the process.

The process of “self-concentration” can be very important in case of ice particles. It is known that ice particles' concentration is highly inhomogeneous with high concentrations often observed in areas of enhanced turbulence.

Summarizing the results of studies dedicated to the problem of droplet spectrum broadening, we conclude that only some aspects of the phenomenon can be explained by suggested mechanisms. For instance, mixing with surrounding leads to formation of small droplets near cloud boundaries while collisions between cloud droplets can be responsible for the formation of cloud droplets, with sizes exceeding the adiabatic value. However, the problem as a whole is not solved yet.
3.6.3.1. Technical notes. The problem of droplet spectrum broadening and the large droplet formation at the diffusion growth stage of cloud development is often artificially avoided in microphysical models in several ways: (a) by prescribing the artificially wide droplet spectrum of nucleated droplets; (b) by introduction of both explicit (using turbulent terms in the equations for size distributions functions) and implicit (numerical viscosity) mixing of droplet size spectra over several neighboring grid points. For example, artificial mixing of droplet size spectra at vertical levels separated by a few hundred meters (a typical resolution of majority of cloud models) leads to artificial arising of smaller droplets at higher levels and larger droplets in the vicinity of cloud base. Horizontal mixing of droplet spectra leads to similar results. It is known that supersaturation and droplet concentration in the vicinity of the cloud base depend on vertical velocity, which attains its maximum at the cloud axis and decreases to the cloud periphery. As a result, the sizes of droplets will be different in different grid points located at the same height level. Mixing of the droplet spectra in the grid points separated in some models by distances of about 1 km will lead to artificial broadening of the droplet spectra; (c) by the employing some algorithms e.g., Kovetz and Olund, 1969 with excessive numerical dispersion of droplet spectra. According to the experience of one of the authors, a decrease in the number of calling Kovetz and Olund’s (1969) procedure by factor of 3 can lead to a delay in rain drop formation by 15–25 min. Liu et al. (1995) suggested a modification of the algorithm for diffusion growth calculation by employing a variational optimization method. A new method significantly reduces numerical diffusion but loses the mass conservation property. Thus, in cloud models the problem is often just opposite of that in the theory: sometimes it is easier to obtain rain in a short time, than to prevent its formation (say in case of a very continental air). We would like to pay attention to the following paradox: the use of adequate schemes which have no numerical broadening can lead (in case a high vertical velocity at the cloud base is assumed) to the formation of extremely narrow, and therefore unrealistic, droplet spectra (Cooper et al., 1997). To obtain the realistic width of droplet spectra in this case, it is necessary to solve the problem of the spectra broadening, taking into account fluctuations of vertical velocity (including those at cloud base), turbulent effects on drop collisions and, possibly, other mechanisms (Korolev and Isaac, 2000).

4. Ice microphysics

The importance of ice phase in clouds was probably first recognized in the early 1930s when Bergeron (1933) hypothesized that ice is required for most heavy rainfall from supercooled clouds. Since that time, much work has been done to further the understanding of the role of ice processes in the precipitation formation, although the role of the solid phase is still plagued with many uncertainties. Many projects of artificial weather modification are based on the assumption that under certain conditions the formation of new ice particles leads to precipitation enhancement.

The effect of cloud ice is not limited to precipitation formation (e.g., Braham, 1964). Cirrus, which are predominantly ice clouds, have a profound climatological effect. The
amount of ice in mixed-phase clouds largely controls the time of beginning and duration of precipitation, radiation properties, and stability of the clouds. In addition, ice particles play crucial role in cloud electrification processes as well as in cloud chemistry.

Despite an important role that ice phase in cloud processes, gaps in our knowledge of ice microphysics are much wider and more significant than in warm rain microphysics.

Although many clouds reside completely or partially above freezing level, cloud droplets do not freeze instantaneously as they are exposed to negative temperatures and can exist in a metastable supercooled condition for a long time. Mixed-phase clouds are common at temperatures warmer than $-20^\circ C$. Liquid droplets have been found in cirrus at temperatures as cold as $-40^\circ C$ (Heymsfield and Miloshevich, 1993). Rosenfeld and Woodley (2000) recently measured 1.9 g m$^{-3}$ of supercooled water at $-37.5^\circ C$, at top of growing cumulonimbus clouds composed of droplets with mean volume diameter of only 17 $\mu$m and concentrations exceeding 900 cm$^{-3}$. Ice formation mechanisms, being of primary importance for cloud evolution, are poorly understood.

4.1. Formation of ice crystals

Ice forming processes are usually divided into primary and secondary categories. In primary processes, also known as nucleation, ice particles are formed directly from liquid water or vapor. Nucleation can be homogeneous, if it occurs when only water substance (vapor and/or liquid) is present, or heterogeneous when the presence of an ice nucleus (IN) is required. Secondary ice production refers to the processes when the formation of new ice crystals involves already existing ice particles. Examples of the secondary processes include the rime-splintering mechanism and ice crystal fragmentation.

4.1.1. Primary ice crystal generation

4.1.1.1. Homogeneous ice nucleation. Drop freezing due to homogeneous ice nucleation is described in terms of a nucleation rate, which denotes the number of liquid-to-solid nucleation events per unit time per unit volume of liquid. The nucleation rate exhibit extremely strong variation with temperature, increasing from $10^{-8}$ cm$^{-3}$ s$^{-1}$ at $-30^\circ C$ to $10^{13}$ cm$^{-3}$ s$^{-1}$ at $-40^\circ C$ (Pruppacher, 1995). Thus, although homogeneous ice nucleation is possible in any supercooled water drop, nucleation rates are negligibly small at temperatures warmer than approximately $-30^\circ C$. Considering the huge range of values, estimated nucleation rates obtained from various theoretical, laboratory, and observational studies during the last decade are in a reasonably good agreement (Sassen and Dodd, 1988; Heymsfield and Miloshevich, 1993; Pruppacher 1995). The remaining uncertainties (of up to an order of magnitude) can be attributed to instrument limitations, inaccuracy of used thermodynamic constants, as well as to variations in chemical composition of cloud water.

The available evidence suggests that homogeneous freezing is the dominant process in the formation of cirrus at temperatures below about $-35^\circ C$ and in tops of deep cumulus forming within a smoky air with a high CCN concentration. Parameterization
of homogeneous droplet freezing for use in mesoscale numerical model with bulk microphysics has been proposed by DeMott et al. (1994).

4.1.1.2. Heterogeneous ice nucleation. Ice forming nuclei (IN) comprise a special subset of the total atmospheric aerosol, similar to cloud condensation nuclei (CCN). Measuring IN concentration remains one of the most challenging problems in experimental cloud physics. Ice nuclei concentration and activity (or size) spectra vary with geographical location, perhaps as strongly as CCN spectra. There is evidence that IN concentration is lower in polar regions than in mid-latitudes (Bigg, 1996). Nevertheless, many more measurements are needed to establish typical values for any specific area. The height dependence of IN concentration is also largely unknown. Fletcher (1962) found little variation of IN count with height in measurement in 1950s. Berezinskii et al. (1986) argued that the relative ice-forming activity of natural aerosol increases with height leading to slower decrease of IN concentration with altitude compared to the well-known decrease in the total aerosol count. In addition to geographical variations of its averaged value, IN concentration anywhere can be affected by synoptic and cloud-scale atmospheric phenomena. Mizuno and Fukuta (1995) documented local changes in IN concentrations, up to an order of magnitude, which they attributed to the passage of cold fronts. An increase in IN concentration as a cold front approached was usually followed by a sharp drop in IN count behind the front, possibly due to precipitation scavenging. Similarly, Al-Naimi and Saunders (1985b) observed a notable reduction in IN concentration near the ground after snow and rain events. Langer et al. (1979) regularly observed 10-fold and larger increases in the IN concentration in the surface outflow of thunderstorms.

The process of ice nucleation is substantially more complicated than the formation of droplets. While CCN are sensitive primarily to the supersaturation with respect to water, the activity of IN depends on supersaturation and temperature. In addition, there are at least four distinct nucleation modes (or mechanisms) through which IN may operate, compared to the one process of activation of CCN. These modes are

(a) deposition nucleation, when water vapor is absorbed directly onto the surface of nucleus where it transforms into ice;
(b) condensation–freezing nucleation, which is a sequence of events when, first a film of liquid is formed on the surface of the nucleus, and then the condensate freezes;
(c) immersion–freezing nucleation, when freezing of droplets is induced by nuclei located within the droplets themselves;
(d) contact nucleation, when drop freezing is caused by the contact of supercooled drops and nucleus.

The theory of the ice nucleation is described in books by Mazin and Shmeter (1983) and Pruppacher and Klett (1997). Below we will concentrate on parameterizations of these mechanisms and their treatment in cloud models. In this section, we consider only the first two modes in which ice crystals are formed on IN from water vapor. The last two mechanisms cause freezing of existing drops and are discussed in Section 4.2.
4.1.1.3. Fletcher’s parameterization. The first quantitative description of average spectrum of ice-forming nuclei in the atmosphere probably should be attributed to Fletcher (1962). He combined data from a dozen of measurement sets by various instruments to arrive to the now famous exponential expression:

$$N_{f1} = N_{f0}\exp\left(-b_{f1}T_c\right)$$  \hspace{1cm} (4.1)

where $N_{f1}$ is the number of nuclei active at temperature warmer than $T_c$ (the Celsius temperature). Note that Fletcher did not distinguish between different nucleation modes. He used measurements conducted within $-15^\circ$C to $-30^\circ$C temperature range and noted large variation of parameters in (4.1). Parameter $b_{f1}$, for example, varied between 0.4 to 0.8, while $N_{f0}$ (with the mean value of $10^{-2} \text{ m}^{-3}$) varied as much as several orders of magnitude between various data sets. It is not clear to what degree these variations are caused by differences in counting methods and techniques, geographical locations, or natural fluctuations in IN concentrations.

Note that (4.1) does not meet the obvious asymptotic condition that at $T_c = 0^\circ$C there should be on IN nucleation. Formula (4.1) is a rather crude parameterization of the combined effects of different processes and modelers must exercise great caution in using it. Detailed criticism of Fletcher’s parameterization can be found in the study by Mazin and Gurovich (1998).

Technical notes. Formula (4.1) has the same characteristic feature as (2.2): it does not explicitly contain time rates of IN activation. The concentration of IN produced in a numerical model by (4.1) depends on the treatment of the formula. In many models, the number of activated IN at each time step is calculated using (4.1), i.e., it is assumed that a certain air parcel contains infinite large source of potential IN, which continue to activate under unchanging conditions. Formula (4.1) being treated in this way predicts an unrealistically large IN concentration (for example, $65 \text{ cm}^{-3}$ at $-30^\circ$C temperature). In addition, the total number nucleated IN in, say 5 min, would depend on the length of the model time step. Consequently, modelers are forced to limit the number of the IN artificially by a maximum value corresponding, for instance, to the temperature of $-27^\circ$C (e.g., Reisner et al., 1998). We suppose that the “overnucleation” is the result of the physically incorrect interpretation of (4.1).

Formula (4.1) should be treated in a “Lagrangian” meaning, similarly to that used in case of (2.2) and (2.7). The maximum value of IN concentration of $65 \text{ cm}^{-3}$ should be understood as the count of IN which becomes active within a certain air parcel during the whole time of rising from the freezing level to the level of $-30^\circ$C. This approach reproduces laboratory conditions, under which the number of active IN increases during air cooling. Thus, expression (4.1) provides a distribution of IN with respect to temperature. The number of IN activated at a certain grid point should be calculated as:

$$dN_{f1} = [-b_{f1}N_{f0}\exp(-b_{f1}T_c)]d(T_c/dt)dt$$

where $d(T_c/dt)$ is the decrease of temperature in an air parcel during its motion toward the grid point during time step $dt$. If $d(T_c/dt) > 0$ there should be no new nucleation: $dN_{f1} = 0$. This approach excludes the “overnucleation” automatically. Recall that in the Eulerian coordinate frame $d(T_c/dt)$ at a certain model grid point can be calculated as $(dT_c/dt) = \frac{\partial T_c}{\partial t} + u(\partial T_c/\partial x) + v(\partial T_c/\partial y) + w(\partial T_c/\partial z)$, where $\partial T_c/\partial t$ is the rate of $T_c$ change in the grid point.
4.1.1.4. Deposition and condensation–freezing nucleation

Theory and observations. Deposition nucleation is a process when water vapor is absorbed directly onto the surface of a nucleus where it is transformed into ice. During condensation–freezing nucleation, a film of liquid is formed on the surface of the nucleus and freezes almost immediately. An important distinction between the two modes is that supersaturation with respect to liquid water is required for condensation–freezing to operate. Under these conditions, however, the deposition nucleation may also take place. Therefore, under water supersaturation condition, it is practically impossible to distinguish between these mechanisms, and a single parameterization is often used to predict the combined effect of the two modes.

Meyers et al. (1992) used data from two sets of continuous-flow diffusion-chamber IN concentration measurements of Rogers (1982) and Al-Naimi and Saunders (1985a,b). The measurements agree to within an order of magnitude for −7°C to −20°C temperature range and for ice supersaturations up to 25%. Based on these data, Meyers et al. (1992) suggested the following formula relating the number concentration of deposition and condensation–freezing IN, \( N \), to the supersaturation with respect to ice, \( S_i \):

\[
N = N_0 \exp\left( a_d S_i \right),
\]

where \( N_0 = 10^{-3} \text{ m}^{-3}, a_d = -0.639, b_d = 12.96. \) This formulation predicts formation of \( 4 \times 10^3 \text{ m}^{-3} \) pristine ice crystals due to deposition and condensation freezing at −15°C under water saturation condition. Nucleation is usually prevented for temperatures warmer than −5°C.

Note that (4.2) does not obey the obvious asymptotic condition that at \( S_i \to 0 \) the number of IN \( N \) should tend to zero.

Expression (4.2) is often used in numerical models for a wider range of conditions than it was originally proposed. The errors induced by such an extrapolation are not known.

Note that the type of activated ice crystals depends on temperature. According to Takahashi et al., 1991

(a) plate-like crystals form at \( -8°C > T_c \geq -14°C \) and \( -18°C > T_c \geq -22.4°C \),
(b) columnar crystals arise at \( -4°C > T_c \geq -8°C, T_c < -22.4°C \),
(c) dendrites (branch-type crystals) form at \( -14°C > T_c \geq -18°C \).

Because crystals of different forms have different growth rates, terminal velocities, and collision efficiencies, it is important to take the temperature effects of crystal formation into account (as it has been done, for instance, by Khain and Sednev, 1996).

Technical notes. Our comments here are similar to those made concerning formulas (2.2) and (4.1). The knowledge of the rate of pristine ice crystal generation is an essential problem of ice microphysics. Adequate evaluation of the time gap between the instance of the maximum supercooled water content and its subsequent elimination by natural ice processes becomes of crucial importance for the appropriate treatment of glaciogenic cloud seeding.
Formula (4.2) actually provides the distribution of number of active IN with respect to supersaturation $S_i$. Modelers “translate” formula (4.2) into rate of the IN concentration using different approaches leading to different results.

Khain and Sednev (1996), Reisin et al. (1996) and Reisner et al. (1998) assume that the IN concentration predicted by (4.2) is a maximum possible value of ice crystal concentration under given $S_i$. If ice crystal concentration in a certain grid point exceeds that predicted by (4.2), new nucleation is prevented. Sometimes the assumption is used that (4.2) provides the maximum number of IN that can be activated a particular grid point. According to our present understanding of the problem, this approach is just intuitive, which has pure physical justification.

Note that the number of the newly activated ice crystals at each time step in a certain grid point, $dN_i$, can be written as:

$$dN_i = \begin{cases} b_d N_i dS_i, & \text{if } dS_i > 0 \\ 0, & \text{if } dS_i < 0 \end{cases}$$

where

$$dS_i = \left( \frac{\partial S_i}{\partial t} + u \frac{\partial S_i}{\partial x} + v \frac{\partial S_i}{\partial y} + w \frac{\partial S_i}{\partial z} \right) dt$$

is an increase in supersaturation in a cloud parcel during its movement from a point $(t-dt, x-dx, y-dy, z-dz)$ to a regular grid point $(t, x, y, z)$ similarly to the case of droplet nucleation (see Eqs. (2.8) and (2.8a)).

From (4.3a), it follows that $dS_i$ should be calculated taking into account advective (convective) terms. The importance of these terms is illustrated in Fig. 12, showing the maximum values of plate-like ice crystal concentration as functions of time during the cloud development in experiment with the ice multiplication excluded. We conducted simulations using an advanced version of the Hebrew University Cloud Model (HUCM), in which three approaches to calculation of IN nucleation were tested: (a) a method based on the assumption that the IN concentration predicted by (4.2) is a maximum possible value of ice crystal concentration under given $S_i$; (b) a method in which advective (convective) terms in (4.3a) are omitted, so that $dS_i = (\partial S_i/\partial t)dt$, is the change in supersaturation in a certain model grid point during one time step; and (c) formulas (4.3) and (4.3a) are used. The time dependence of maximum liquid water content is also shown. One can see that these approaches provide different rates of ice crystal generation. The maximum rate is reached in case when $N_i$ is referred to as a maximum possible value of ice crystal concentration existing under a given $S_i$ and immediate nucleation of $N_i$ ice crystals during one time step is assumed (in case of no nucleation in the particular grid point earlier). Until the ice multiplication is not included, the utilization of $dS_i = (\partial S_i/\partial t)dt$ leads to a significantly lower concentration of ice crystals. Use of (4.3a) leads in our case to intermediate rate of ice crystal formation. At early stage, (4.3a) leads to the lower rate of ice crystal generation as compared to the case, when immediate nucleation of $N_i$ number of ice crystals during one time step is assumed (with time delay of about 5–6 min). Thus, the use of (4.3a) leads in our case to significant increase in the time gap during which a large content of
Fig. 12. Maximum values of plate-type ice crystal concentration as functions of time during the cloud development according to Meyers et al. (1992) formula when (a) an approach, in which concentration of ice crystals according to this formula is interpreted as the maximum possible value, nucleated immediately (curve $N_{\text{max}}$), and (b) a “Lagrangian” approach is used (curve $dS/dt$); (c) the partial time derivative of supersaturation is used for calculation of the nucleation rate (curve $dS/dt$). The time dependence of maximum liquid water content (in g m$^{-3}$) is also shown (curve LWC).

supercooled water coexists with a very low concentration of pristine ice crystals. At $t > 40$ min, the maxima of ice crystal concentration obtained using these two approaches turn out to be similar and greater than that in the case when only $dS/dt$ is used. Note that these three approaches result in significant difference of spatial distributions of ice crystal concentration (not shown). The difference follows from different rates of ice generation (both in time and space) under the same cloud dynamics (primary ice generation does not influence the velocity field).

We believe that the Lagrangian approach, expressed by formulas (4.3) and (4.3a), is a natural physical approach, based on laboratory experiments. According to laboratory experiments, the number of IN within a certain air probe increases by $dN_i$ in case supersaturation increases by $dS_i$. We suppose, therefore, that in case this volume experiences the change of supersaturation $dS_i$ during its motion in a cloud, a number of new activated IN within this volume will be equal to $dN_i$ (we assume that characteristic time of IN activation is less than that of the transport of IN between model grid points).

We believe that the problem of ice crystal generation should be solved similarly to that in case of CCN nucleation in warm microphysics, namely, by implementation of a budget (and distributions) of IN into models. As soon as the size distribution of IN is given, the sink of IN by activation will decrease the number of IN available. The Lagrangian approach seems to be a reasonable way to describe the sink of IN. We suppose that the next step in development of treatment of ice generation should be implementation of IN distributions. To solve the problem, a better understanding of the nature and properties of IN is required.
Approaches based on the introduction of some constraints, maximum and minimum values of variables (say, IN concentration), etc., can provide results suitable for many simulation purposes. However, these approaches do not describe the physics of processes. In addition, measurements indicate the existence of remarkable variations of ice crystal concentration. Therefore, sometimes, it is not clear what values should be preset to be realistic in particular points of a cloud.

4.1.2. Secondary ice crystal production

In the cloud modeling community, the Hallet and Mossop (1974) rime-splintering mechanism remains the most widely used ice multiplication process. Hallet and Mossop (1974) and Mossop and Hallet (1974) found that this phenomenon occurs under a narrow range of conditions. At a temperature of $-5^\circ C$, one splinter (ice crystal of the smallest resolvable size) is created for every 200 droplets with diameter greater than 24 $\mu$m collected by riming graupel. This maximum splinter production rate decreases toward the ends of the temperature interval, which are set approximately at $-3^\circ C$ and $-8^\circ C$. The question of whether the splinter production rate should be related to the ambient air temperature (Mossop, 1985) or the rimer surface temperature (Heymsfield and Mossop, 1984) is likely to remain open until the exact physical mechanism of splinter production is determined. The precise mechanism of splinter formation, which is often referred to as the Hallet–Mossop process, is not yet known although several hypotheses have been put forward (e.g., Mossop, 1976; Griggs and Choularton, 1983; Dong and Hallet, 1989; Mason, 1996).

The process of ice multiplication is rather effective. It starts, however, only after graupel particles appear in a relatively high concentration.

To account for the rime-splintering rate correctly, concentration of droplets with diameters greater than 24 $\mu$m and smaller than 13 $\mu$m must be known. This presents a problem for bulk-type microphysical models where the shape of the droplet spectrum is usually fixed. In such models, the number concentration of large (or small) droplets is directly and monotonically related to the liquid water content, meaning that the large-drop tail becomes important only for spectra with high liquid water content. This is not the case in real clouds or in bin-resolving models where the width of the droplet spectra is variable and bimodal spectra are commonly found.

Although the Hallett–Mossop mechanism is the best-documented secondary ice production process, other ice multiplication processes have been suggested. Vardiman (1978) proposed an empirical relation relating the number of splinters produced in an ice–ice collisions to the change in momentum of interacting particles. Takahashi et al. (1995) suggested formation of secondary ice particles in graupel–graupel collisions. It has also been suggested that fragmentation may occur during ice crystal evaporation. Recent laboratory studies confirm ice crystal breakup during evaporation (Swanson et al., 1998). Limited amount of data available so far prevents the development of any general parameterization of these effects.

4.2. Drop freezing: contact and immersion nucleation

There are two main mechanisms of heterogeneous drop freezing contact freezing and immersion freezing (Pruppacher and Klett, 1997).
4.2.1. Contact freezing (contact nucleation)

Unlike deposition/condensation–freezing IN, the presence of active contact IN does not automatically lead to formation of ice particles. For a supercooled water drop to freeze, it must collide with a contact IN. Brownian diffusion, thermophoresis, diffusiophoresis, and inertial impaction prompt these collisions. In order to take into account the contact freezing, wet scavenging of aerosol has to be modeled.

It should be noted that in addition to all difficulties of measuring concentration of natural ice nuclei in general, evaluation of concentration of contact nuclei represents even a greater challenge. The reason for additional uncertainty is that nucleation rate of contact mode crucially depends upon nucleus size, which is not known and is difficult to measure. Assuming different sizes and, therefore, different efficiencies of various capturing mechanisms, one would deduce values of ice nucleus concentrations that vary by orders of magnitude and, yet, are based on the same data set. For example, using Blanchard’s (1957) data and assuming contact nucleus size of 0.3–0.5 \( \mu \)m, Young (1974) deduced concentration of \( 10^3 \) to \( 10^6 \) m\(^{-3}\) for nuclei active at \(-4^\circ\)C. Deshler and Vali (1992) re-evaluated the Blanchard’s experiment assuming nucleus radius of 0.05 \( \mu \)m and found that the required concentration reduces to \( 2 \cdot 10^4 \) m\(^{-3}\) for the same temperature. Beard (1992) came to a similar estimate for even smaller nucleus radius of 0.01 \( \mu \)m. He also showed that if the assumed size is increased to 2 to 5 \( \mu \)m, only in the order of \( 10^2 \) m\(^{-3}\) of such giant nuclei is needed to explain the observed freezing rate.

Meyers et al. (1992) analyzed nearly all available data and offered a parameterization by fitting a function to the measurements of Vali (1976), Cooper (1980), and Deshler (1982):

\[
N_{\text{ca}} = N_{c0} \exp(a_c - b_c T_c),
\]

where \( N_{\text{ca}} \) is the number concentration of contact nuclei active at temperature \( T_c \) (in \(^\circ\)C), \( a_c = -2.8; \) \( b_c = 0.2629 (\circ\)C\(^{-1}\)), \( N_{c0} = 10^3 \) m\(^{-3}\). No data exist for concentration of contact IN at temperatures warmer than \(-10^\circ\)C. Thus, any extrapolation of the dependency (4.4) into this temperature range is speculative. At \( T_c = -11^\circ\)C formula (4.4) gives concentration of contact IN of \( 10^3 \) m\(^{-3}\).

Note that because of (1) the counts of active IN activated according to (4.2) and (4.4) are of the same order, and (2) the collision efficiencies of the scavenging are very small, the concentration of ice particles generated by the contact freezing is by a factor of about \( 10^3 \) smaller than that generated by the deposition/condensation freezing (Ovtchinnikov, 1998). There is a significant difference between deposition–condensation and contact freezing. While in the first case only small ice crystals are formed, in the second case large frozen drops can be produced. Thus, the effect of contact IN depends not only on concentration, size and other parameters of contact IN, but also on the size distribution of unfrozen drops.

Note that the variability of \( N_{c0} \) in real clouds due to variations in ambient aerosol concentrations is not known, but can be quite high. Thus, under certain conditions, the concentration of ice particles (frozen drops) formed by the contact freezing can be greater than the above estimate.

Although some of the cloud models are capable of adequate simulation of scavenging process (Respondek et al., 1995; Ovtchinnikov and Kogan, 2000; Ovtchinnikov et al.,
2000), parameterization of contact nucleation remains a serious problem due to lack of reliable data on nucleus concentration and size.

4.2.1.1. Technical notes. In some models (e.g., Khain and Sednev, 1996; Reisin et al., 1996) formula (4.4) is used along with (4.2) without distinguishing the physical nature of processes, i.e. \( N_{in} \) is considered as a number of nucleated ice crystals. It leads, possibly, to some overestimation of pristine ice formation rate in these models. This error is, however, much less significant (taking into account that \( N_{in} \) and \( N_{d} \) are the same order) than the error resulting from incorrect treatment of (4.2), as it was discussed above.

4.2.2. Immersion freezing

Immersion IN are immersed in droplets and activate their freezing under supercooled conditions. It has long been discovered that the mean freezing temperature of drops depends on their volume (or mass) and perhaps the rate of cooling (e.g., Bigg, 1953; Pitter and Pruppacher, 1973). It is usually assumed that immersion INs are distributed homogeneously throughout liquid cloud water. Thus, the larger drops have larger probability of containing an immersion nucleus. A theory of immersion nucleation is not yet complete. There are two main hypotheses upon which two types of parameterizations are based.

The stochastic hypothesis treats immersion freezing in a way analogous to homogeneous freezing. It assumes that the freezing results from a random formation of a critical size embryo, and the presence of foreign particles increases the probability of the nucleation without disturbing its stochastic nature. Many models use immersion–freezing parameterizations based on the stochastic hypothesis formulated by Bigg (1953) (e.g., Reisner et al.—MM5 model; Khain and Sednev, 1996—the HUCM; Reisin et al., 1996). Following Bigg (1953), the freezing probability is assumed proportional to drop mass and the freezing rate is calculated as:

\[
\frac{1}{f_w} \frac{\partial f_w}{\partial t} = -a_t m \exp\left(-b_t T_c\right),
\]

where \( f_w \) is the size distribution of water drops, \( m \) is the drop mass, \( a_t = 10^{-4} \text{ s}^{-1} \text{ g}^{-1} \), \( b_t = 0.66 \text{ (°C)}^{-1} \) (Wisner et al., 1972; Reisner et al., 1998). From formula (4.5) the time needed to freeze one half of the existing drops with mass \( m \) can be calculated as:

\[
t_m = \left[ \ln 2 / (a_t m) \right] \exp(b_t T_c).
\]

This time \( t_m \) rapidly decreases with height above the freezing level. For instance, at \( T_c = -20°C \) and \( m = 4 \cdot 10^{-3} \text{ g} \) (\( r = 1000 \text{ μm} \)), \( m = 4 \cdot 10^{-6} \text{ g} \) (\( r = 100 \text{ μm} \)) and \( m = 4 \cdot 10^{-9} \text{ g} \) (\( r = 10 \text{ μm} \)) the values of \( t_m \) are 2 s, 2 \( \cdot 10^3 \) s and 2 \( \cdot 10^6 \) s, respectively. At \( T_c = -30°C \), the values of \( t_m \) are of 2 \( \cdot 10^{-3} \) s, 2 s and 2 \( \cdot 10^3 \) s, respectively. These evaluations show that according to formula (4.6) rain drops freeze at warm temperatures such as at \(-10°C\), while small cloud droplets can exist for a comparably long time at temperatures as low as \(-30°C\). Nevertheless, their freezing leads to the production of enormous amount of small ice crystals.
Although at temperatures warmer than $-10^\circ\text{C}$ even large drops have low probability of freezing, such events may be important because they provide instantaneous riming centers and could trigger the rime-splintering multiplication process.

Note that expression (4.6) has been deduced from laboratory experiments with distilled water. Such water contains solid particles which, although numerous, are relatively small and uniform in composition. This is in contrast to the case of cloud water, which contains a strongly heterogeneous mixture of AP. It is therefore not surprising that the freezing spectra of drops made from cloud and rainwater is quite irregular in comparison to that from distilled water (Pruppacher and Klett, 1997). Formula (4.6) provides much stronger temperature dependence than was measured for cloud and rainwater.

At temperatures colder than $-20^\circ\text{C}$, parameterization (4.5) produces ice crystal concentrations that are usually attributed to ice multiplication.

An alternative to the stochastic hypothesis is the singular hypothesis, which assumes that the drop freezing temperature is determined by nucleus properties (Vali, 1994). Unlike the stochastic hypothesis, the number of frozen drops under singular parameterization does not change with time for a given supercooling. It is assumed that immersion nuclei are distributed homogeneously throughout liquid cloud water and that their activity increases with decreasing temperature. A parameterization based on the singular hypothesis is also consistent with two experimentally determined tendencies that more drops of a given size freeze at colder temperatures, and larger drops are more likely to freeze at a given temperature. Ovtchinnikov and Kogan (2000) used a temperature dependence of immersion nuclei given by Vali (1975):

$$ N_{im} = N_{im0}(0.1 T_f)^\gamma $$

where $N_{im}$ is a number of active inversion nuclei per unit volume of liquid water, $N_{im0} = 10^7 \text{ m}^{-3}$, $\gamma = 4.4$ cumuliform clouds.

Formula (4.7) reveals much lower temperature dependence of active immersion IN than (4.5). For instance, the number of active IN within the temperature range from $-20^\circ\text{C}$ to $-30^\circ\text{C}$ increases by factor 1000 in case of (4.5) and only by a factor of about 7 in case of (4.7). Use of (4.7) instead of (4.5) makes the immersion freezing less effective than the contact freezing in production of ice particles at temperatures above $-15^\circ\text{C}$ (Ovtchinnikov, 1998).

Although the actual drop freezing is probably better represented by some combination of these mechanisms than by either one acting alone (Pruppacher and Klett, 1997), current models use either stochastic or singular approach. Vali (1994) proposed a model for a time-dependent freezing rate that may serve as a basis for an improved parameterization of immersion freezing in cloud models. The main limitations of the immersion–freezing parameterizations arise from inadequate knowledge about the freezing nucleus content in the cloud water, which is difficult to obtain from either theory, or from direct sampling.

4.2.2.1. Technical notes. Note, that formula (4.5) cannot be considered as adequate for the complete range of temperatures. For example, an evident condition of no freezing
when $T_r \to 0$ is not satisfied. To obey the obvious boundary conditions, the expression $\exp(-b_u T_c) - 1$ is to be used instead of $\exp(-b_u T_c)$.

4.3. Other mechanisms of ice formation

It has been suggested that evaporating cloud droplets may provide an additional source of IN. There is only circumstantial evidence that cloud droplet residues may serve as ice nuclei (Rosinski and Morgan, 1991; Beard, 1992). It is even less clear in which mode such nuclei would operate. However, evaporation nuclei may provide an attractive explanation for observed ice enhancement. First, this mechanism appears to be fairly general, since droplets evaporate in any cloud in great numbers. Secondly, high IN concentrations are often observed in downdrafts and in mature and eroding turrets, which are also the locations favorable for the formation of evaporation nuclei. Finally, the activity of these nuclei may be enhanced because of the electrical charge accumulated on their surface during evaporation (Beard, 1992). More needs to be learned about rates of production, active modes, and size spectra of evaporation nuclei before a meaningful parameterization can be constructed for use in cloud models.

Czys (1989) presented an experiment in which the freezing of 3-mm drops was stimulated by a mechanical shock. Based on the estimation of collision pressure, Czys speculated that collision of supercooled drops in clouds might result in freezing only when drizzle-size drops are present. No quantitative parameterization of this mechanism has been proposed and more experimental and theoretical studies of this effect are required.

4.4. Deposition and sublimation of ice

Growth rate of ice particles with mass $m_i$ by water vapor deposition/sublimation can be expressed as (Pruppacher and Klett, 1997):

$$\frac{dm_i}{dt} = aC_i\left(S_i - \frac{b}{C_i} Q\right),$$

(4.8)

where coefficients $a$ and $b$ depend on temperature, thermal conductivity, diffusivity and other properties of air, as well as on relative ice-particle–air velocities (through so-called ventilation coefficients). $Q$ is the additional heat source (e.g., due to freezing of liquid water accumulated during riming), $C_i$ is the capacitance of the crystal. For a spherical particle, $C_i$ is equal to the radius.

Capacitance of different types of ice particles is determined by rather complicated formulas, containing geometrical parameters of crystals. The differences are significant and lead to different growth rates of ice particles of different type. Moreover, the form (e.g., aspect ratio) of ice particles of a certain type changes during their growth, so that $C_i$ is the function of not only the type, but also the size of the particle.

We know only one example of a cloud model (Khain and Sednev, 1996; Khain et al., 1999a) where differences in capacitance of different ice crystals (plate, columnar and dendrites), as well as effects of particle form changes during particle growth, are taken into account. These effects lead to different composition of cloud ice and influence radiative cloud properties.
In most models, the effect of heating on ice particle growth is neglected. This effect becomes important, however, when hail growth/evaporation is simulated.

4.5. Collection growth of ice particles

4.5.1. Types of collisions

Collection growth of ice particles is one of the most complicated problems in mixed-phase microphysics. It is related first of all to great number of interactions between particles of different kind. An important difference of ice–ice and ice–water interaction from drop–drop collisions is that the resulting particle may be of a type different from the type of parent particles. For instance, it is reasonable to assume that coalescence of ice crystals of different type leads to snowflake formation.

As an example, we present here some “rules” used by Khain and Sednev (1996) in case of single acts of particles collisions (see the paper for more detail):

(a) Drop–drop collisions: new particles are water drops.
(b) Drop–crystals’ collisions: if the mass of drops is less than that of crystals new ice crystals are formed, otherwise either graupel or hail is formed depending on air temperature.
(c) Drop–snowflakes’ (aggregates’) collisions: if the mass of drops is less than that of snowflakes new ice snowflakes are formed, otherwise either graupel or hail is formed depending on air temperature.
(d) Drop–graupel collisions: either graupel or hail is formed depending on air temperature.
(e) Drop–hail collisions: the result is hailstone formation.
(f) Ice crystal–ice crystal: snowflakes are formed.
(g) Ice crystal–snowflake (aggregates): snowflakes are formed.
(h) Snowflake–snowflake: snowflakes are formed.
(i) Graupel–ice crystal and graupel–snowflake collisions: graupel forms if its mass is larger, otherwise, crystal or snowflake is formed.

We define graupel as rimed ice particles with density ranging from 0.1 to 0.8 g cm$^{-3}$. Khain and Sednev (1996) and Khain et al. (1999a) set graupel density equal to 0.4 g cm$^{-3}$. Hail/frozen drop particles are defined as ice particles with the density equal to 0.9 g cm$^{-3}$. Note that these rules are valid for single acts of particle collisions. Continuing use of these rules for drop–ice collisions can lead to artifacts. For instance, in the model, a large ice crystal can collect a great number of water drops while remaining an ice crystal. In reality, the rimed fraction (total mass of collected drops) after a certain number of collisions with drops can become so significant that the crystal should be interpreted as graupel. To treat this transition correctly, the rimed fraction of ice particles must be known. The perfect solution would be to calculate the densities of rimed particles and to redistribute ice particles between different types based on their densities. The problem is rather complicated, taking into account a large number of collisions between particles of different kind. In a new version of HUCM (Khain et al., 1999c), it is assumed that rimen of ice crystals and snowflakes leads to graupel formation in case the mass of rimed water exceeds half of particle mass.
The process of collisions in a mixed-phase cloud is described by a system of stochastic kinetic equations for all types of cloud hydrometeors. These equations look like Eq. (2.9), but are complicated by a great number of interactions taken into account (see, e.g., Khain and Sednev, 1995). As in the case of drop–drop collision, the rate of collisions is determined by the collision kernels, which describe the probability of particles moving with different velocities to collide. The collision kernels are determined by the collision efficiency and swept volume, both of which depend on the form of the colliding particles and their relative velocity. As was previously noted, the collisions between different cloud particles influence the microphysical composition of a mixed-phase cloud. Thus, the knowledge of the collection kernels is of great importance.

4.5.2. Riming

Ice precipitation forms mainly by drop–ice collisions, i.e., by process of riming.

When water drop–ice collisions are considered, one needs to distinguish between the cases when ice particles serve as collectors of small droplets, and when large drops collect ice particles.

In the first case, some data concerning ice crystals–droplet collision efficiency is available (Pruppacher and Klett, 1997). It is known that there is a minimum threshold size (a cutoff value) of ice crystals, below which they are unable to collect small droplets. This minimal size is about 100 μm for plates and about 50 μm for columnar crystals, respectively (Pruppacher and Klett, 1997). It is likely that ice crystals of other shapes also have similar threshold sizes. According to the diffusion growth equation of ice crystals (Rogers and Yau, 1989), the characteristic time needed for plate crystals to reach radius of 100 μm under the values of supersaturation with respect to ice of 20% (typical value found at temperatures of −15°C to −20°C in the HUCM, Khain and Sednev, 1996) can be evaluated as 1000 s. Thus, small ice crystals, even in high concentration, are unable to decrease significantly the cloud water content in short amount of time by collisions.

In some studies claiming to explain rapid cloud glaciation (e.g., Ryan, 1973) the collision efficiencies between small droplets and small crystals were highly overestimated. Even under these conditions the existence of significant amount of raindrops was necessary to simulate cloud glaciation during a few tens of minutes (we suppose that the process of ice crystal growth at the expense of evaporating drops can be one of the important mechanisms of rapid glaciation (Swarzenbock et al., 1999)).

The efficiency of snowflakes–droplet collisions and coalescence are poorly known. It is strongly affected by the porous effects, which depend on the snowflake structure, temperature and other parameters. Porous effects seem to increase the coalescence efficiency by a factor of 5 to 10, especially for very small droplets and AP. This is one of the reasons for high scavenging ability of snowflakes (Pruppacher and Klett, 1997).

The efficiencies of ice-crystal collection by larger water drops are also largely unknown. To our knowledge, there are only a few studies where the collision efficiencies between drops of several hundred micron in radius and plate-like and columnar crystals are presented (Lew and Pruppacher, 1983; Lew et al., 1985). The lack of data forces modelers to choose values of the collision efficiencies based largely on their intuition.
Because of the lack of the appropriate experimental data, many modelers assume that the collision efficiency of graupel and water drops is equal to the collision efficiency of water drops of the same size (e.g., Beheng, 1978). In some other studies, the assumption that the graupel–drop collision efficiency is equal to the collision efficiency of water drops of corresponding masses is used (e.g., Khain and Sednev, 1996; Khain et al., 1999a). In the latter case, the graupel–drop and drop–drop collision kernels turn out to be close: graupel has larger size, but lower terminal fall velocity.

Johnson (1987) carried out a quantitative comparison of the continuous riming growth rates for graupel with those of unfrozen drops growing by coalescence. He showed that graupel had an advantage over unfrozen raindrops in the regions where cloud droplets were comparably large (e.g., 20 \( \mu \text{m} \) in diameter). This difference was reached mainly due to smaller coalescence efficiency of water drops (the fraction of collisions that results in coalescence), which was taken from empirical formulas of Beard and Ochs (1984). Johnson assumed that the coalescence efficiency of graupel–drop collisions is equal to 100%, while the coalescence efficiency of drop collisions was ranged from 60% to 80%. The graupel–drop and drop–drop collision efficiencies were assumed equal in this study as well.

At the same time, it is known that the crystal–drop collision efficiencies differ significantly from those of drop–drop collisions (Pruppacher and Klett, 1997). We mentioned above that there is a cut-off size of ice crystals (including crystals having the form close to spherical) below which ice crystals are unable to collect small water droplets. At the same time, drop–drop collisions are always possible in a pure gravity case.

The assumption that the graupel–drop collision efficiencies are equal to the collision efficiencies of drops is based on the intuitive conception that the collision efficiency is determined by the shape of colliding particles only. This intuitive assumption is incorrect: the collision efficiencies are determined not only by the shape of colliding particles, but depend on the relative velocity between particles. Because the densities and, consequently, terminal velocities of graupel (and other ice particles) are not equal to those of water drops of the same mass (or size), the graupel–drop collision efficiencies must be different from the collision efficiencies of drops. Detailed hydrodynamical calculation of graupel–drop collisions have been conducted recently by Khain et al. (1999b) under the assumption that graupel are spherical particles of different densities.

It was shown that at low (0.1 g cm\(^{-3}\)) and middle (0.4 g cm\(^{-3}\)) densities, a graupel collector has significantly lower collision efficiencies with cloud droplets than a drop-collector of either the same size or mass. As an example, we present Fig. 13, which shows the graupel–water collision efficiencies for 0.4 g cm\(^{-3}\) graupel density. The collision efficiencies of water drops are presented as well to facilitate comparison. Similarly to ice crystals–drop collisions, the graupel–drop collision efficiency increases with droplet size from zero to a maximum value and then sharply decreases to zero, when the terminal velocity of droplets approaches that of graupel. As soon as drop terminal velocity becomes higher than that of graupel (so water drops capture graupel) the collision efficiency experiences a jump to the values significantly exceeding one, and then decreases quickly to about one with the increase of drop size. This asymmetry in
the collision efficiency behavior can be attributed to the differences in the velocity fields induced by graupel and drop falling with similar velocities. Because of larger size, the graupel is characterized by larger Reynolds number than the corresponding drop. Consequently, mutual tracks of the particles during their hydrodynamic interaction depend on whether graupel or drop serves as a collector and located above.

The problem becomes more complicated, if one takes into account that ice particles have complex shapes, so that particles of the same have different terminal velocities. Tracks of non-spherical particles can be complicated even in the calm atmosphere. Drop–ice relative velocities and the droplets and ice collision kernels depend on orientation of ice particles of ice particles, which in its turn depends on the flow velocity.

The problem of determining the collection efficiency as a function of temperature, humidity and other parameters also remains largely unsolved.

Results illustrated in Fig. 13 were obtained under calm air conditions (a pure gravity case). We expect, however, a significant increase of the graupel–drop collision kernels in turbulent clouds. First, graupel form in clouds at the mature stage when the turbulence intensity is significant and the dissipation rate can be above $1000 \text{ cm}^2 \text{ s}^{-3}$, i.e. by order of magnitude higher than at earlier stage of cloud development (about $100 \text{ cm}^2 \text{ s}^{-3}$) (Panchev, 1971; Mazin et al., 1984; Weil et al., 1993). The latter value was used by Pinsky et al. (1999c) for evolution of turbulence effects on the collision efficiency of
small droplets. According to Pinsky et al. (1998b), in case of high intensity turbulence, the swept volume for middle density graupel with radii ranged from 300–500 μm can be by a factor of 4 larger than that in pure gravity case. The corresponding increase for a drop-collector is smaller by a factor of 1.5. Based on these evaluations, Pinsky et al. (1998b) concluded that the growth rate of graupel by accretion in a highly turbulent flow could be higher than that of drops.

Note that Pinsky et al. (1998b) draw their conclusion by a comparison of continuous growth rates of graupel and drop-collectors under the assumption of the equality of the graupel–drop and drop–drop collision efficiencies. The calculation of graupel–drop collision efficiencies in a turbulent flow is the subject of further investigations. As soon as the results are obtained, the comparison of efficiency of coalescence and riming in real clouds will be possible.

4.5.3. Ice–ice collisions

Ice–ice collisions determine the rate of snowflakes formation and snow precipitation. Uncertainties in the problem are even more pronounced than those in case of water–ice collisions. There are very few data concerning the collisions of ice crystals (Pruppacher and Klett, 1997). The values of collection efficiencies reported by different authors vary by two orders of magnitude (from $10^{-3}$ to $10^{-1}$). The collision efficiency depends on the shape and densities of ice particles. Coalescence efficiency depends on the properties of ice particles surfaces, temperature, and humidity. Khain and Sednev (1996) assumed that the collection efficiencies are close to zero at temperatures close to $-25^\circ$C, while at $0^\circ$C they were assumed equal to those between drops of equivalent sizes. The dependence on the supersaturation with respect to ice is also assumed. Additional laboratory studies are required to clarify the problem.

It should be noted that the turbulent effects in case of ice–ice collisions must be much more pronounced than in case of drop–drop collisions. Pinsky and Khain (1998) showed that due to ice particle inertia, volumes swept by ice crystals during their motion in a turbulent flow significantly increased. The contribution of turbulence effects on the swept volume can be characterized by the ratio of the RMS value of the relative velocity between particles induced by their inertia $\langle \Delta V^2 \rangle^{1/2}$ and the relative velocity induced by gravity $\Delta V = V_{i2} - V_{i1}$. In cases when the ratio is much less than one, the turbulence effects can be neglected. If the ratio is greater than one, the relative velocity between particles is formed mainly due to turbulence effects. Fig. 14 shows the dependence of the ratio $\langle \Delta V^2 \rangle^{1/2} / \Delta V$ on the sizes of colliding particles for (a) collisions between plate crystals and (b) collisions of plate crystals and water droplets. The dissipation rate $\varepsilon$ was set equal to $100 \text{ cm}^2 \text{ s}^{-3}$. One can see that the magnitudes of $\langle \Delta V^2 \rangle^{1/2} / \Delta V$ for the smallest particles reach maximum values. Even at comparably low turbulence intensity, these values can be as large as two. Thus, it is possible that the relative velocity between ice crystals is determined mainly by turbulence and not by gravity. Comparison of values $\langle \Delta V^2 \rangle^{1/2} / \Delta V$ for ice particles with the corresponding ratios for water drops shows that in the case of ice particle, the ratios retain their high values for much larger sizes than in the case of water drops.

The problem requires further investigation, as significant turbulence effects on the ice–ice collision efficiency are also expected.
Fig. 14. Dependence of ration \( \langle \Delta V^{1/2} \rangle^{1/2} / \Delta V \) on the bulk sizes of ice particles of different type about to collide for the dissipation rate \( \varepsilon = 100 \text{ cm}^3 \text{ s}^{-1} \).
4.6. Melting

During melting, liquid water and ice co-exist on the same particle. Multidimensional cloud models are unable to treat adequately the mixed-phased particles. The crudest parameterization is that of instantaneous melting at the freezing level when all ice particles are transformed into water drops of corresponding masses. Deficiency of the simplification can significantly affect cloud dynamics as well as microphysics, especially when large ice particles are present and an extensive part of the cloud is below the freezing level. An alternative is to calculate the melted fraction of particle mass and to distribute it among drops of specified sizes. This approach requires a priori assumptions about the size of shed droplets for which only little information is available (Rasmussen and Heymsfield, 1987).

Raindrops resulted from melted graupel and snowflakes can be rather large, and despite their low concentration, produce high radar reflectivity. Many studies are dedicated to this problem, and its analysis is beyond the framework of this review.

5. An example of cloud–aerosol interaction effects on cloud microphysics

Many cloud models, as well as cloud resolving mesoscale models have been applied to investigate different environmental problems related to precipitation formation, rain chemistry, effects of cloud microphysics on radiative cloud properties (e.g., Bott, 2000), effects of cloud seeding on rain amount, etc. Cloud–aerosol interaction is increasingly recognized as one of the key factors controlling precipitation regime on local, meso- and even global scales.

One of the main mechanisms through which atmospheric AP influence cloud development and precipitation formation is the process of droplet nucleation, when APs serving as CCN give rise to the formation of new droplets. The concentration of nucleated droplets depends on concentration, size and chemical composition of APs. High concentration of nucleated droplets slows down droplet growth by diffusion and the sizes for the process of drop collisions to become efficient cannot be reached. Recent observations of cloud development within different types of air masses (maritime or continental type) reveal crucially different cloud development and rain formation. Rosenfeld and Lensky (1998a,b) and Lensky and Rosenfeld (1998) observed that in polluted areas over Thailand and Indonesia, smoked clouds do not precipitate altogether, having narrow spectra of small droplets. At the same time, similar clouds precipitate in clear air in only 15–20 min after their formation. Kaufman and Nakajima (1993) found significant decrease droplet size (from 15 to 9 μm) accompanied by an increase in drop concentration in continental clouds of Amazon smoked area. Martinsson et al. (1999) found significant increase of concentration (up to 2000 cm$^{-3}$) together with a substantial reduction of effective droplet radius in polluted orographic clouds over a mountain ridge. Pawlowska and Brenguier (1998) found strong dependence of maritime stratocumulus microstructure upon the aerosol background in precise measurements. Recent observations made with the Tropical Rainfall Measuring Mission (TRMM) satellite data
demonstrated that smoke from burning vegetation can practically shut off warm rain formation in tropical clouds (Rosenfeld, 1999). These observations indicate that aerosols influence precipitation regimes on meso- and possibly global scales. The impact of aerosols on the rainfall is, supposedly, one of the most important issues of anthropogenic climate change (e.g., Hobbs, 1993).

The high sensitivity of rain rate and amount in cumulus clouds to the concentration and size of AP was reported in numerical simulations using advanced spectral microphysics models (e.g., Reisin et al., 1996; Khain et al., 1999a; Yin et al., 2000). Significant effect of aerosols on microphysics of stratiform clouds was found by Liu and Kogan (1999) using a large eddy simulation numerical model. Bradbury et al. (1999) numerically found significant modification of aerosol resulting from passage through a hill cap cloud.

An example below illustrates the important role of aerosol concentration on cloud droplet spectrum and rain production. This example also highlights the necessity of a proper treatment of microphysical processes in cloud models.

In several numerical experiments with the HUCM (Khain et al., 1999a), the cloud development in the Eastern Mediterranean coastal zone during a cold season was simulated when the sea surface is warmer than the land surface by about 5°C. Low-level wind convergence between dominating westerly wind and the coastal breeze-like circulation creates favorable conditions for development of convective clouds over the sea about 20 km offshore. The clouds are transported by the background flow inland. Initial vertical profiles of AP concentration were assumed exponential with maximum values at the surface 130 cm$^{-3}$ (exp. E1) and 1000 cm$^{-3}$ (exp. E2). Fig. 15 shows vertical profiles of the CWC, rain water content (RWC) and liquid water content (LWC) in the zone of active convection in these two experiments at 1 h. One can see that CWC in E1 (maritime air) is significantly lower than in E2 (polluted air). However, RWC in E1 (maritime air) is more than twice as large as in E2 (polluted air). This is because drop concentration in E2 is several times higher than in E1 (not shown) and, therefore, cloud droplets in E2 are significantly smaller than in E1.

In Fig. 15 we also see that rainwater forms in the polluted air at higher levels than in maritime air. In case of polluted air, significant content of supercooled water remains at levels with temperature as low as $-30^\circ$C (which agrees with recent observations by Rosenfeld and Woodley, 2000), while in the maritime air there is no significant LWC at levels with temperature as high as $-15^\circ$C. Thus, cloud–aerosol interaction impacts crucially the cloud microphysics via the influence on the droplet spectrum width.

The difference in drop size and locations of maximums of unfrozen raindrops in cases of low and higher AP concentration leads to significant difference in ice microphysics as well.

5.1. Technical note

In some bulk microphysics mesoscale models (e.g., Pielke et al., 1992; Reisner et al., 1998) the autoconversion rate (the rate of rain drop formation as a result of cloud droplets collision) is described by the following formula (Kessler, 1969): $A_u =$
max\{ K(CWC − CWC_{\text{min}}), 0 \}, where CWC_{\text{min}} is an empirical threshold value. K is the empirical constant. This equation ignores any dependency of raindrop production on the cloud droplet size and width of the droplet size spectrum. The latter is the major controlling factor of rain formation. Comparison of the CWC and RWC in E1 and E2 shows the rate of precipitation formation can differ from that predicted by the Kessler formula by factor of 5 to 10! The Kessler formula can predict incorrectly even tendency in the rain drop generation with increase of AP concentration.

Note that the RAMS model no longer uses Kessler’s formula, but instead employs lookup tables of autoconversion rates generated from bin model computations based on the assumed bulk distribution functions (Feingold et al., 1998).

Fig. 15. Vertical profiles of the cloud water content (CWC) and rain water content (RWC) and CWC and liquid water content (LWC) in the zone of active convection in two experiments with different aerosol concentration (τ = 1 h) (see text for more details).
6. Conclusions

We have presented a review of the state-of-the-art modeling of some microphysical processes in clouds. Because of the complexity and broadness of the subject, the review does not claim to present an exhaustive analysis of the status in the field.

In the review, we focused on two problems: (1) gaps in our knowledge of the microphysical processes, and (2) the ways of representation of those microphysical processes, mathematical descriptions of which (parameterizations) are considered to be known from a theory or observations.

The gaps in our knowledge are significant. The efforts in future investigation should be directed to cover these gaps. The particular problems to be solved can be summarized as follows.

The main theoretical problems in warm rain processes seem to be an adequate simulation of droplet spectra formation. This problem requires the following points.

(a) The development of a general theory of droplet formation and growth by diffusion. Broadening of the spectrum can be attributed to in-cloud nucleation of new droplets through the process of supersaturation growth in excess of the cloud base maximum and to the mixing of cloud air with drop-free environment. The formation of supersaturation in excess over the cloud base maximum can be caused (a) by acceleration of vertical velocity induced by buoyancy, (b) by turbulent fluctuations of vertical velocity, (c) by fluctuations of droplet concentration caused by turbulent including inertia effects mechanisms, (d) by mixing of air volumes of different temperatures, etc. All the effects suggest a crucial impact of both “dynamic” and “aerosol” components on droplet spectrum formation.

Further development of physically and mathematically grounded approaches intended for the description of the complicated process of mixing in a turbulent flow, as well as of the turbulent flow structure at high Reynolds numbers, are needed.

(b) An adequate treatment of droplet collisions taking into account turbulent effects, which seem to increase significantly the probability of cloud droplets coalescence. Height dependence of the collision efficiency should also be taken into account.

(c) Determination of the role of giant CCN in rain formation. Reliable observations of concentration of giant CCN under different conditions are required.

We see the main problems of the ice-phase microphysics in the following points.

(a) The description of processes of ice formation in clouds must be improved. The formulas used so far are some kind of parameterizations based on laboratory measurements or obtained by averaging the results of observations under different conditions. We suppose that the problem of ice crystal generation should be solved similarly to the of CCN nucleation in warm microphysics, by implementation of budget and distributions of IN into models. As soon as the size distribution of IN is given, the sink of IN by activation will decrease the number of IN available. We suppose that the next step in the development of treatment of the ice generation should be the implementation of IN distributions. To solve the problem, a better understanding of IN is required.

(b) The role of the process of ice particles growth at the expense of evaporating droplets is not well understood (in particular, when cloud glaciation is investigated) and poorly simulated in numerical models.
(c) Lack of data on the collection rates of ice particles due to drop–ice and ice–ice collisions. The gaps in these data do not permit us to evaluate adequately the role of ice processes in mixed-phase clouds. Turbulence effects should be investigated and taken into account. Recent results concerning the turbulent effects and increase of the collision rates with height indicate that the collision rates of ice particles are possibly underestimated so far.

(d) Ice microphysics (ice concentration, amount and type) depends on the spectra of water drops lifting above the freezing level. It means that ice-phase microphysics is determined largely by cloud–aerosol interaction and the processes of droplet spectrum formation. Thus, appropriate treatment of cloud–aerosol interaction is one of the key problems of both warm and mixed-phase microphysics.

Analysis of approaches used to simulate cloud and rain formation in cumulus clouds using bin (spectral) microphysics cloud models shows the following.

(a) Despite the fact that dynamic effects (the rate of atmosphere instability, air humidity, vertical velocity) were well known, they were understood mainly in a “quantitative” sense, as a general influence, say, on total cloud and rainwater contents. Recent theoretical studies indicate that thermodynamic conditions influence droplet spectra formation through cloud–aerosol interaction. Thus, the knowledge of dynamic parameters such as vertical velocity and its distribution at the cloud base, CCN concentration and distribution, vertical gradients of temperature in the cloud layer, etc., turn out to be basic parameters determining cloud and rain formation processes.

The utilization of arbitrary initial temperature/humidity pulses (as well as other types of initial forcing) triggering cloud development can lead to an incorrect droplet spectrum development in case these conditions result in unrealistic vertical velocity at the cloud base. The cloud-boundary layer interaction determining the distribution of vertical velocity at the cloud base and aerosol composition turns out to be one of the central problems. Different cloud parcels ascending from the cloud base with different velocities from different droplet spectra. Because the sizes of individual cloud parcels is usually smaller than resolution usually used in cloud models, the problem of parameterization of their effects on the microphysics of whole clouds arises.

(b) Some cloud models produce rain because of artificial droplet spectrum broadening caused by numerical schemes with significant “numerical viscosity” of schemes of diffusion and collision drop growth. The rate of the artificial droplet spectrum broadening depends on the number of mass bins and some other factors. It is paradoxical that just a decrease in time steps can increase the rate of artificial droplet spectrum broadening, because processes leading to this broadening will be “called” (used) more often. One of important problems is to eliminate artificial factors and implement physically grounded mechanisms responsible for cloud and precipitation formation.

Analysis of representation of microphysical processes in numerical models shows that even in those cases, when parameterization expressions of different processes are given, the ways they are implemented in cloud models are different, sometimes questionable or incorrect. This leads to the necessity to introduce artificial assumptions, limits, and parameters to keep results within a reasonable range of values. Applying certain artificial bounds of variations of model variables reflects actually lack in understanding of corresponding processes. This remark is especially relevant to implementation of
experimental (laboratory) expressions, which do not include time explicitly. Different treatments of the same parameterization lead to significantly different results. Hence, the majority of references to formula suggested by, say, Fletcher (1962), Bigg (1953), Vali (1975), Meyers et al. (1992), and others will provide little information unless the way of their implementation is described. Based on our experience, we have recommended several approaches that proved their precision and efficiency in cloud modeling.

We limit our review only to some aspects of cloud microphysics and their simulations using spectral (bin) microphysics models. Some important problems remained beyond the frame of the paper. For example, a widely used method of model integration is splitting with respect to physical (microphysical) processes. In real clouds these processes take place at the same time. In models they are treated in a certain order. In case the time steps are much smaller than the characteristic time scales of different microphysical processes, the order chosen, supposedly, does not play an important role. However, the time scale of many microphysical processes is smaller than the time steps of several seconds usually used for model integration. In this case, the order chosen can influence the results of integration. We suppose that when the splitting with respect to microphysical processes is used, the treatment of these processes should take into account their characteristic time scales, beginning with the most rapid processes.

The requirements imposed on the precision of one or another model are determined by the purposes to be attained and the problems to be solved. In order to predict the intensity and spatial distribution of precipitation (including heavy rain, hail, and aggregates) over several kilometer size regions, a high precision when treating microphysical processes is required. This precision is especially important in some agricultural areas, urban zones, complex terrain zones, where floods are possible, etc.

It is interesting that the microphysical methods based on the first principles are required for simulations of climate and climate change. This is because simulations of possible scenarios of climatic changes require the utilization of certain parameterization parameters, which can differ from the values chosen for the present climatic conditions.

Cloud–aerosol interaction plays a crucial role in precipitation formation and influences thermodynamic (for example, vertical profiles of latent heat release) and radiative cloud properties, and hence, the climate and climate changes. Thus, cloud models should properly simulate the processes of cloud–aerosol interaction. Results of advanced models indicate that AP formed by biomass burning can eliminate precipitation even from deep tropical clouds over large areas. The effects of aerosol on the climate may be even more significant than those related to changes in the atmospheric ozone. Thus, along with model improvement, a corresponding measurement system permitting modelers to simulate aerosol effects should be developed.

Especially high precision is required in simulations of cloud seeding effects. For instance, the existence (or the lack) of the gap between formation of large unfrozen drops and the formation of a large amount of ice can determine, possibly, the success or failure of glaciogenic seeding hypothesis. The same comment is valid with respect to hygroscopic seeding, the efficiency of which is determined by the width of the cloud drop spectrum and by the rate of the spectrum broadening under natural conditions. Thus, realistic simulations of cloud seeding effects impose strict demands on the precision and sophistication of cloud microphysical models.
Successive reproduction of these processes and events can be attained on the way of the better understanding of microphysical processes in clouds.

Acknowledgements

The authors are grateful to Prof. I. Mazin for valuable comments and remarks. This study was partially supported by the Germany–Israel Science Foundation (grant 0407-008.08/95), and by the Israel Science Foundation founded by the Israel Academy of Sciences and Arts (grant 572/97) and the Israel Ministry of Science (grant 84291-96) and the Environmental Sciences Division of the U.S. Department of Energy (through Pacific Northwest National Laboratory Contract 144880-A-Q1 to the Cooperative Institute for Mesoscale Meteorological Studies).

References


A. Khain et al.

Atmospheric Research 55 2000 159–224


