The role of competition effect in the raindrop formation

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Abstract

Large cloud condensational nuclei (CCN) affect droplet spectrum evolution in two ways: first, droplets growing on these nuclei are larger than others and accelerate the collision of cloud droplets having larger collision kernel; second, the condensation growth of large soluble aerosols between cloud base and the level of supersaturation maximum decreases the magnitude of the maximum which prevents activation of the smallest or less soluble nuclei and decreases the concentration of nucleated droplets. The latter process is known as competition effect. This effect is usually assumed to be an important factor leading to the rain enhancement by hygroscopic seeding.

This study tests the hypothesis that the magnitude of the supersaturation maximum is reduced significantly when large CCN are present. A 2000 bin 1-D cloud parcel model was used to test this hypothesis. The model calculates diffusion aerosol growth and corresponding decrease in air relative humidity both below and above cloud base. It explicitly simulates droplet nucleation without any parameterization. The model calculates droplet concentration at the level of the supersaturation maximum under different concentrations and sizes of large soluble nuclei. Collisions are calculated by solving a stochastic kinetic equation for collision.

The results of our investigation indicate that the previous studies have overestimated the efficiency of competition effect. The reasons of the overestimation of the effect in earlier studies are presented. The main effect of large soluble aerosols is the formation of large droplets which accelerate droplet collisions.

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1. Introduction

Renewed interest in hygroscopic seeding for rain enhancement has come as a consequence of three recent experiments reporting a statistically significant increase in rainfall from individual clouds (Mather et al., 1996, 1997; Bigg, 1997; Bruintjes, 1999; Silverman and Sukarnjanaset, 2000). However some unresolved issues remain concerning the mechanisms by means of which seed particles lead to the rain enhancement. These issues are closely related to the effect aerosol particles have on the droplet spectrum formation.

The size of nucleated droplets depends on size and chemical composition of aerosol particles. Since the main purpose of a study is limited by the reproduction of the condensational growth of these nuclei, the simplified description is possible (“pure physical” approach). According to this approach nuclei with different solubility are characterized by an “equivalent” radius of completely soluble nuclei that produce droplets of the same size as actual aerosol particles (Mazin and Shmeter, 1983). Similarly, we replaced the size distribution of actual aerosols with a corresponding size distribution of completely soluble aerosol particles.

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of the equivalent size. In this approach droplets growing on larger cloud condensational nuclei (CCN) attain larger sizes than other droplets. Note in this connection that in most cases (especially for maritime aerosols) the equivalent radii are quite close to the corresponding radii of real aerosols (Mazin and Shmeter, 1983; Khain et al., 2000).

We distinguish activated CCN and non-activated CCN following terminology used by Pruppacher and Klett (1997). This terminology allows us to use the term “CCN” even in those cases when the term “condensation nuclei (CN)” is commonly used.

Diffusion growth of fully soluble particles and their activation into droplets depend on their size. Large CCN (defined here as particles belonging to so called “coarse aerosol mode” activated at supersaturation below 0.004% (that corresponds to NaCl particles of the radii >0.5 μm, Rogers and Yau, 1989) affect the droplet spectrum evolution in two ways: first, droplets growing on these CCN accelerate the collision process (e.g., Segal et al., 2004a); second, the diffusion growth of large CCN decreases supersaturation preventing activation of the smallest CCN. The latter mechanism is known as “the competition effect”. The decrease of the droplet concentration accelerates droplet growth by diffusion and fosters the raindrop formation by auto conversion. The role of the competition effect has been discussed in literature for a significant period of time. Gerard and Clement (1978) have stated that “Competition between hygroscopic seeding particles and natural condensation nuclei should be minimal due to relatively low density of seeding particulates which are anticipated to be required (on the order of 1 cm–3)”. However, in recent studies regarding hygroscopic seeding, the concentration of seeded particles required to achieve a significant rain enhancement was estimated as high as ~1 cm–3 (or 103 l–1) and higher (Cooper et al., 1997; Segal et al., 2004b). Such concentrations are no longer negligible. For instance the mass of 1 μm-radius particle may be of the same order of magnitude as the mass of all natural nuclei with the mean radius of 0.1 μm-radius. Respectively a significant role of the competition effect on the droplet concentration is often assumed (e.g., Bruintjes, 1999).

The purpose of this study is to quantitatively evaluate the competition effect, using a 2000 bin cloud parcel model (Pinsky and Khain, 2002; Segal et al., 2004a,b). This model is chosen since it explicitly outputs nuclei growth and the corresponding decrease in the air relative humidity both above and below cloud base. The simulations were performed under different thermodynamic conditions typical of intermediate (Mediterranean) and continental environmental (Texas) conditions.

2. Model description

The simulations are carried out using a 2000-bin spectral microphysics cloud parcel model (1-D model) allowing turbulent effects on droplet collisions (Pinsky and Khain, 2002). The model equation system includes (e.g., Pruppacher and Klett, 1997): the diffusion growth equation used for nuclei and water droplets, the stochastic kinetic equation for droplet collisions, equations for supersaturation, vertical velocity and the temperature of the ascending parcel. The acceleration of cloud parcel velocity is determined by the buoyancy (including liquid water loading) and the drag force. The drag force is assumed to be proportional to the square of the updraft velocity of the parcel. The proportionality parameter λ = 0.2/R is assumed to be inversely proportional to the cross-sectional radius R of a parcel. These drag equations are well acknowledged (e.g., Pruppacher and Klett, 1997).

The model contains a precise description of warm rain microphysics. Non-activated nuclei and cloud droplets are described by a mass grid containing 2000 mass bins within the radius range of 0.01 μm–2000 μm. The mass of each bin in the mass grid changes with time (height) according to the equation of diffusion growth. No special simplifying approach to distinguish non-activated CCN and droplets are applied. The separation of sizes and growth rates between droplets and non-activated aerosols (haze particles) is reproduced automatically when the largest aerosol particles start rapidly growing as droplets or continue growing toward their equilibrium size, while smaller particles remain to be in equilibrium with the environment. This Lagrangian approach eliminates the artificial spectrum broadening typical of models that use unmovable regular mass grids. The precise method proposed by (Bott, 1998) is used to solve the stochastic collision equation. Collision droplet growth was calculated using a collision efficiency table with a unique high resolution in droplet radius of 1 μm (Pinsky et al., 2001). The effects of turbulence on the collision kernels were taken into account (Khain et al., 2000; Pinsky and Khain, 2002).

The values of velocity and accelerations of cloud parcel are determined by environment temperature profile, as well as by the drag force (through the entrainment parameter λ). By varying parameter λ we simulated parcels reaching different maximum levels under the same thermodynamic environment.
The time step of 0.001 s is used to calculate the diffusion growth of drops and aerosol particles. Such a small time step is necessary to simulate adequately the growth of the smallest nuclei, so that separation between non-activated nuclei attaining equilibrium and the growing droplets is simulated explicitly (without parameterization) by solving the equation for the diffusion growth. Drop growth by collisions is calculated using a time increment of 0.5 s.

As a rule, drop sedimentation is not taken into account in cloud parcel models. Nevertheless, the present model includes calculation of the liquid water mass flux (referred to as rain flux), as well as of the total rain amount that falls down from an ascending cloud parcel (per square unit). These calculations are conducted as follows. Using the values of terminal fall velocities of droplets of different mass, as well as the velocity of parcel updraft, the flux of drops leaving a cloud parcel during a unit of time is calculated. The flux, integrated all over the period of the cloud parcel ascent, provides the evaluation of the total rain amount falling down from the parcel. It is clear that the precipitation rate and accumulated rain in a cloud consisting of many cloud parcels (cells) can differ from those following from this approach. Nevertheless, this approach characterizes the raindrop production rate in ascending air and is useful in experiments aimed at comparing droplet spectrum evolution under different aerosol size distributions.

As it discussed by Pinsky and Khain (2002), the model can be regarded as benchmark method as concerns the reproduction of supersaturation and droplet concentration near the cloud base. This model was used for the development of a parameterization of droplet concentration as the function of concentration and size distribution width of CCN and cloud base velocity (Segal and Khain, in press). This parameterization allowed reproduction of the dependences observed in different field experiments (Ramanathan et al., 2001).

2.1. Design of numerical experiment conditions

Ascending cloud parcels were simulated under two thermodynamic conditions: (a) Typical of the Eastern Mediterranean rainy season (Fig. 1a). This sounding reveals 75% relative humidity near the surface, where the temperature is equal to 17 °C. (b) Unstable continental condition (Fig. 1b), exemplified by the Midland/Texas sounding data of 13 August 1999 (Rosenfeld and Woodley, 2000; Khain et al., 2001). This sounding reveals 35% relative humidity near the surface, where the temperature is equal to 36 °C.

The initial spectrum of dry CCN is represented by a superposition of three modes of the log-normal distribution (Hobbs et al., 1985, Respondek et al., 1995) as:

$$\frac{dN}{d\ln r_n} = \sum_{i=1}^{3} n_i \frac{1}{\sqrt{2\pi} \sigma_i \ln 10} \exp \left( -\frac{(\log_{10} r_n - \mu_i)^2}{2(\log_{10} \sigma_i)^2} \right)$$

where $n_i$ is the maximum number concentration in the $i$th mode, $R_i$ and $\sigma_i$ are the mean radius and the width of the $i$th aerosol mode. CCN of small and intermediate size are represented by first two modes with $R_1=0.006 \mu m$, $\log(\sigma_1)=0.3$ and $R_2=0.03 \mu m$, $\log(\sigma_1) =0.3$, respectively. The distribution (1) was used successfully by Segal et al. (2004a,b) for simulation of clouds under the Mediterranean and Texas conditions. The first mode represents nuclei with radii $<0.01 \mu m$ (the number of particles exceeding this size in the mode is negligible). The smaller nuclei remain typically haze aerosols in clouds with moderate vertical velocities. The nuclei belonging to the second mode represent the main source for cloud droplets. They represent largest particles in the “nuclei” mode and the “accumulation” nuclei mode. The third mode represents large CCN which activated at supersaturation $<0.004%$ belonging to the coarse aerosol mode. The distribution (1) changes in the ascending air parcels due to the CCN activation (droplet nucleation).

In our calculations it was assumed that aerosols consisted of NaCl. In case the chemical composition of aerosol particles differs from NaCl, one can easily calculate “effective” size distribution of NaCl particles having the nucleation properties, similar to those of the non-NaCl particles (Mazin and Shmeter, 1983). Indeed, the nucleation properties of the aerosols are determined by “chemistry” term in the equation of diffusion growth, which contains the product $Br_N^3$, where $B$ is the coefficient responsible for the chemical properties of aerosols (it includes the Van’t Hoff factor, the molecular weight and dry bulk density), and $r_N$ is the radius of the dry aerosol particle. The radius of equivalent NaCl particle can be calculated as $r_N^{NaCl} = r_N(B/B_{NaCl})^{1/3}$. Note that the ratio $(B/B_{NaCl})^{1/3}$ is often quite close to one because of the power (1/3). It is especially true for maritime aerosols that contain a significant fraction of NaCl aerosols (e.g., Clarke et al., 2003).

In control simulations no CCN belonging to the third mode were included in the CCN distribution. The competition effect is investigated by adding large CCN to the CCN spectra in the starting point of parcel ascent. Large CCN were assumed to have a very narrow
distribution. We repeated simulations using mean radius of large CCN between 0.5 μm and 10.0 μm and varying the concentration from 0.125 to 256 cm$^{-3}$. In each simulation vertical profile of supersaturation and droplet concentrations were calculated. The competition effect is evaluated by the concentration of large CCN required to decrease the droplet concentration by a given percent (1% and 10%). These large CCN concentrations will be referred to as critical ones.

Cloud parcel ascent from the surface was triggered by small temperature fluctuations. Typically, the updraft velocities at cloud base were about 1 m s$^{-1}$ in the Mediterranean case, and 3 m s$^{-1}$ in the continental cases. In control simulations the droplet concentration at the level of supersaturation maximum (a few tens meters above the cloud base) was 520 cm$^{-3}$ and 1050 cm$^{-3}$ under the Mediterranean and continental conditions, respectively. These values are typical of clouds developing under these conditions (Rosenfeld and Woodley, 2000; Khain et al., 2001; Segal et al., 2004a).

The results are presented in Sections 3.1 and 3.2. In Section 3.1 we discuss the effect of competition on the

Fig. 1. Soundings typical of the Eastern Mediterranean rainy season (upper panel), and typical of continental conditions exemplified by the Midland/Texas sounding data of 13 August 1999 (low panel).
droplet concentration. In Section 3.2 the effect of the concentration decrease on the raindrop formation and on the accumulated rain is discussed. Possible reasons of overestimation of the competition effect in earlier studies are discussed in Section 3.3. Conclusions are presented in Section 4.

3. Results

3.1. Evaluation of competition effect on droplet concentration

3.1.1. Sensitivity to changes in nuclei size

The efficiency of the competition effect will be evaluated in terms of the concentration of large CCN required to decrease the concentration of nucleated droplets by a certain percent (for instance by 10%). This concentration will be referred to as critical one. A size dependence of the critical concentration of large CCN required to decrease the concentration of cloud droplets by 1% and by 10% under different thermodynamic conditions is shown Fig. 2. One can see that the decrease of nucleated droplets concentration by 10% requires \( \sim 10 \) times higher concentration of large CCN as compared to that required to decrease droplet concentration by 1%. The critical concentration of large CCN depends on the CCN size. A 10% decrease in droplet concentration could be attained by “seeding” with 0.4 \( \mu \text{m} \)-radii CCN dispersed at about 20 CCN \( \text{cm}^{-3} \), or by “seeding” with 10 \( \mu \text{m} \)-radii CCN dispersed at 2–3 CCN \( \text{cm}^{-3} \). Note that such high concentrations of large CCN rarely exist under natural conditions, but can be, possibly, formed under seed conditions.

Sensitivity of the competition effect to the concentration of small CCN was also investigated. This involved varying the concentration of CCN in the second CCN spectrum mode between 100 \( \text{cm}^{-3} \) and 2000 \( \text{cm}^{-3} \). Segal et al. (2004b) reported results of hygroscopic cloud seeding simulations using the same parcel model. Seed CCN of different radii were tested. It was found that the seed reagent consisting of large particles with dry radius of 2.5 \( \mu \text{m} \) (referred to as the optimum radius) leads to the maximum raindrop mass production at a given mass of the reagent (consisted of NaCl). We also investigated the competition effect assuming the mean radius of large CCN was 2.5 \( \mu \text{m} \). For sake of comparison the simulations with mean radius of 1 \( \mu \text{m} \) radius CCN were performed as well. Fig. 3a,b show relationship between concentration of large CCN required to decrease the concentration of nucleated droplets by 1% and 10% for maritime and Texas conditions. One can see that the critical concentration of large CCN is actually independent of the small CCN concentration because the mass of the water vapor absorbed by these small particles during their activation is small and hardly affect supersaturation value.

3.1.2. Sensitivity to changes of vertical velocity

An increase in droplet concentration can be attained in two different ways: by an increase in the CCN concentration and by an increase in the vertical velocity. The increase in the vertical velocity leads to an increase supersaturation at cloud base and to activation of smaller CCN, which remain non-activated at lower updrafts. The effect of vertical velocity at cloud base was investigated by changing it from 0.6 to about 7.1 \( \text{m s}^{-1} \) (Fig. 4). The

![Fig. 2. Dependence of the critical concentration of large CCN on their size under Mediterranean (intermediate) and Texas (continental) conditions. The critical CCN concentration is evaluated as the concentration needed to decrease the drop concentration by 1% or by 10%.](image)
variation of the vertical velocity was attained by variation of initial temperature in air parcels at the initial level near the surface. These results are useful for further discussion of the effect of competition caused by large CCN.

Dependencies of the critical concentrations of large CCN on cloud base velocity obtained in simulations are shown in Fig. 5a,b. One can see that the increase in the concentration of droplets at the cloud base caused by the increase in velocity leads to a substantial increase the critical concentration of large CCN (i.e. to decrease the competition efficiency) both under the Mediterranean (Fig. 5a) and the Texas (Fig. 5b) thermodynamic conditions. The difference in the results presented in Fig. 5a,b from those shown in Fig. 3a,b can be explained as follows. The increase in the parcel velocity leads to an increase in supersaturation and to the activation of larger number of CCN. Thus, to decrease the droplet concentration by the same percent under higher vertical velocity a higher the concentration of large CCN is required. Thus, the efficiency of the compensation mechanism decreases with the increase of cloud base updraft.

3.2. The effect of competition on the raindrop production

(Figs. 2, 3 and 5) show that a significant concentration of large CCN is required to decrease droplet concentration by a comparatively small percentage. It

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**Fig. 3.** (a) The critical concentration of 1.0 \( \mu m \) and 2.5 \( \mu m \)-radii CCN needed to decrease the concentration of nucleated droplets by 1.0% and 10% vs. the droplet concentration at cloud base. Simulations were performed under Mediterranean thermodynamic conditions. (b) Similar to (a), but for clouds developing under continental thermodynamic conditions.
Fig. 4. The concentration of nucleated droplets at cloud base of parcels developed under the Mediterranean and Texas thermodynamic conditions as the function of the cloud base velocity.

Fig. 5. (a) Dependence of the concentration of 2.5 μm-radii CCN required for the decrease of the concentration of nucleated droplets by 1% and 10% at different vertical velocities at cloud base. (b) Similar to Fig. 5a, but for the Texas thermodynamic conditions.
will be shown below that the decrease in the droplet concentration that can be attained by the competition is not the major factor that affects raindrop production. To justify the statement two sets of simulations were performed. The first set simulations were performed with different concentrations of CCN belonging to the second aerosol mode. No large CCN belonging to the third mode was allowed in these simulations. In each simulation droplet concentration and parcel accumulated rain were calculated. In the second set of simulations the 2.5 μm-radius nuclei (the optimal size) were added to the aerosol spectrum. The concentration of these large nuclei was varied from 0.025 cm$^{-3}$ to 25.6 cm$^{-3}$. Our simulated decrease in the droplet concentration was caused by the presence of large CCN. Comparison of accumulated rain attained under similar droplet concentrations in these two simulation sets allows revealing the dominating mechanism affecting rain formation in an ascending cloud parcel (effects of droplet concentration decrease-pure competition effect, and the presence of large CCN accelerating droplet collisions.

Results of these simulations performed under the Mediterranean thermodynamic conditions are presented in Fig. 6a–d. The simulations were carried out with cloud parcels of different size attaining different cloud top heights. The results show that a decrease in droplet concentration in simulations with no large CCN leads to a quite small (about 1%) increase in the parcel accumulated rain. The same decrease of the droplet concentration caused by large CCN indicates much higher rain enhancement. Results shown in Fig. 6a–d indicate, therefore, that rain enhancement is caused by the effect of large CCN on droplet collisions, but not by

Fig. 6. (a) Accumulated rain in a parcel of 1650 m depth vs. the concentration of nucleated droplets at the cloud base. The curve marked by filled circles represents results obtained in case with the no large CCN in the CCN size spectrum. The curve marked by open circles represents the cases where 2.5 μm-dry radii CCN were assumed in the spectra near the surface. Numbers in the figures represent concentration of these large CCN. (b) Similar to (a), but for a cloud parcel reaching 3250 m height. (c) Similar to (a), but for a cloud parcel reaching 4200 m height. (d) Similar to (a), but for a cloud parcel reaching 5500 m height.
the decrease in the concentration of small droplets (i.e. not via the competition effect).

Fig. 6a–d indicate a non-linear dependence of the parcel accumulated rain on the concentration of large CCN. The increase in the concentration of 2.5 μm-radii CCN from zero up to \( \sim 1.6 - 2 \times 10^3 \) cm\(^{-3}\) leads first to a very rapid rain enhancement. Further increase in the large CCN concentration leads to much slower increase in the raindrop production, because the 2 cm\(^{-3}\) concentration of large CCN allows realization the cloud parcel precipitation potential.

Note that these conclusions are obtained in neglecting ice processes. Ice processes affect precipitation formation significantly, so that accounting for these processes may change these evaluations.

3.3. Factors influencing the competition effect

Results of calculations indicate that the competition effect is ineffective as concerns the decrease of droplet concentration. This conclusion contradicts the common opinion about the efficiency of this mechanism. We believe that the opinion comes from a simple evaluation of amount of water vapor that large particles can absorb and a comparison of this amount with the value of supersaturation maximum at cloud base. Indeed, the amount of water vapor that can be condensed on large CCN particles is quite significant. For instance, according to the calculations the radius of initially dry 2.5 μm-radius CCN at the level of supersaturation maximum is about 12 μm. Assuming the concentration of these dry 2.5 μm-radius nuclei at 1 cm\(^{-3}\), one can evaluate the water mass condensed by these nuclei as \( \sim 10^{-2} \) cm\(^{-3}\). Given a cloud base mixing ratio of about 5 g m\(^{-3}\), the water condensed onto these large CCN is estimated to be 0.2% of the cloud base (i.e. saturated) mixing ratio. Since the supersaturation maximum near cloud base is close to 0.2% (Fig. 8), one could expect that the competition effect is significant, at least in case of high concentration of large CCN.

More detailed analysis indicates that this simple evaluation is not correct. Soluble aerosols grow below cloud base at relative humidity exceeding humidity of deliquesce \( S_{\text{del}} \) (e.g., 75.3% for pure NaCl nuclei). Most large CCN do not attain their critical size (that exceeds 20 μm) (Rogers and Yau, 1989), i.e. remain formally to be non-activated aerosols. The nuclei of other chemical composition (e.g. consisted of ammonium sulfate) reveal similar behavior. Fig. 7 shows vertical profiles of “wet” radii of large CCN with dry radius of 2.5 μm under different concentrations of these CCN. The particles were “injected” into air parcels near surface. Calculations show that the CCN particles of 2.5 μm attain radii of 11–11.5 μm at the cloud base level. Keeping in mind that the radii of these particles at the supersaturation maximum level is 12–12.5 μm, we conclude that large CCN get the major fraction (about 2/3) of their mass below cloud base, and only 1/3 of their mass within the narrow cloud layer below the supersaturation maximum level. Since the magnitude of the supersaturation maximum is affected by diffusion growth of CCN above cloud base only, the effect of large CCN on the supersaturation maximum turns to be quite small (Fig. 8). Thus, large CCN decrease the cloud base supersaturation maximum much less than it follows.
from the assumption that the major increase in large CCN mass takes place within a cloud.

However, the decrease in the supersaturation maximum is even smaller than that following a simple evaluation of the water vapor mass condensed on the large CCN within this cloud layer. The matter of fact, the supersaturation in an ascending parcel is determined by the following equation (e.g., Pruppacher and Klett, 1997; Pinsky and Khain, 2002):

\[
\frac{dS}{dt} = (S + 1)A_1 W - A_2 \frac{dq}{dt},
\]

where \( S \) is the supersaturation, \( W \) is the vertical velocity, \( q \) is the CWC, \( A_1 \) and \( A_2 \) are the coefficients slightly dependent on temperature. The second term on the right hand of the equation describes the loss of supersaturation due to the water vapor condensation on droplets and growing hygroscopic aerosols. One can see that the changes in the supersaturation are determined not only by the second term, but also by the first one. An increase in the concentration of large CCN (e.g., by seeding) leads to the increase of both terms on the right hand. The first term increases due to increase in \( W \) caused by the corresponding increase in the latent heat release (the latter leads to an increase in the depth of the layer between cloud base and the level of supersaturation maximum) and some other factors.

Even in the case of extremely high concentration of large nuclei (10 cm\(^{-3}\)), the supersaturation maximum decreases only by \( \sim 30\% \) (from 0.35\% to 0.25\%). Below we present the budget estimations leading to this result.

Expression (2) can be written in the integral form being integrated from the cloud base height (\( S=0 \)) to the level of supersaturation maximum. With a high accuracy, this “budget” equation can be written in the form: 
\[
\Delta S = S_{\text{max}} - A_1 \Delta z - A_2 \Delta q,
\]

where \( \Delta z \) is the distance from cloud base to the level of supersaturation maximum, \( \Delta q \) is the increase of condensed water within this layer. According to model calculations in case of no large CCN (“no seeding”), the magnitudes of the terms \( S_{\text{max}} \), \( A_1 \Delta z \) and \( A_2 \Delta q \) are 0.0035, 0.0054 and 0.0019, respectively. In case of seeding with the 2.5 μm-radius CCN with concentration of 10 cm\(^{-3}\), the magnitudes of these terms turned out to be 0.0025, 0.01 and 0.0075, respectively. One can see that \( S_{\text{max}} \) decreases by 30\% from 0.35\% to 0.25\%. This comparatively small decrease can be attributed to the fact that a significant increase of the second term \( A_2 \Delta q \) from 0.0019 to 0.0075 is largely compensated by the increase in the first term \( A_1 \Delta z \). The increase in \( \Delta z \) is seen in Fig. 8.

There are some additional factors that weaken the competition effect. Note first that condensation of water vapor on nuclei increases buoyancy and vertical velocity of ascending air parcel (Fig. 9). As a result, cloud base velocity in the presence of large CCN is somehow larger leading to an increase in supersaturation and the concentration of nucleated droplets.

Second, large CCN decrease the relative humidity in the subcloud layer leading to a corresponding increase in the cloud base level (Fig. 8). Since velocity of ascending parcel increases with height in our simulations, the increase of the cloud base height leads to a
further increase of cloud base velocity (in addition to the effect of latent heat release). To evaluate quantitatively these effects, we performed supplemental sensitivity simulations, in which (a) no decrease in water vapor mixing ratio by water vapor condensation on large CCN was allowed. This condition prevents the change of the cloud base level; and (b) no effects of latent heat release on the vertical velocities were allowed. In these simulations vertical velocity was assumed equal to that calculated in corresponding simulations with no large CCN. The results of these sensitivity experiments are shown in Fig. 10. The figure represents the dependence of critical concentration of large CCN (required to decrease the droplet concentration by a certain percentage) on the radius of these CCN. The results of simulation in which all effects are taken into account are referred to as natural, the simulations in which no effect of large CCN on the mixing ratio and no effect on vertical velocity were taken into account are denoted as $\Delta(dq/dz) = 0$ and $\Delta(dw/dz) = 0$, respectively. One can see...
that if no thermodynamic and dynamic effects of large CCN are taken into account, the critical concentration of these large CCN decreases 2–3 times. It means that neglecting thermodynamic and dynamic effects of large CCN below cloud base leads to an additional overestimation of efficiency of the competition effect several times.

Note that the results of calculations presented in Figs. 7 and 8 indicate the maximum effect of large CCN on the supersaturation (and on droplet concentration). For instance, the seeding with dry (i.e. smaller size) CCN performed at the cloud base would result in a much smaller effect, because (as it follows from the equation of the diffusion growth) the mass growth rate of CCN and droplets increases with their size.

Note that the conditions applied in the evaluations presented in the beginning of the section and in supplemental runs are commonly used in numerical simulations of aerosol effects on clouds, as well as in cloud seeding simulations. For instance, most models do not include aerosols growth (and corresponding thermodynamic effects) below cloud base. The growth of the aerosols is assumed to be at the expense of water vapor within a cloud layer below the supersaturation maximum. This assumption drastically overestimates the competition effect efficiency.

4. Conclusion

The effect of large soluble aerosols (natural or seeded) on droplet concentration at cloud base and on rain drop formation is examined. The analysis is performed using a 2000 bin spectral parcel model with detailed microphysics. An important model feature is the calculation of the diffusion growth of the aerosol particles below cloud base, as well as corresponding latent heat release and decrease of the air humidity.

Simulations were performed for cloud parcels developed under intermediate (Mediterranean) and continental (Texas) thermodynamic conditions.

It was found that the droplet concentration at cloud base decreases with an increase in concentration and size of large CCN. However, a very high concentration of large CCN is required to decrease the droplet concentration even by 10%. It was also found that the competition effect depends on concentration of small CCN only slightly, because of low effect of small CCN on the air humidity. A 10% decrease in droplet concentration could be attained by “seeding” with 0.4 μm-radii CCN dispersed at about 20 CCN cm⁻³, or by “seeding” with 10 μm-radii CCN dispersed at 2–3 CCN cm⁻³.

According to results obtained, the effect of competition on the cloud base droplet concentration is significantly smaller than it was believed earlier. The increase of the CCN mass below cloud base affects the cloud base level only. The additional mass growth of large CCN within cloud from the cloud base to the supersaturation maximum is relatively small, so that it affects the supersaturation maximum only slightly.

The decrease in the supersaturation maximum turns out even smaller than that following from a simple evaluation of the water vapor mass condensed on the large CCN within the cloud layer from the cloud base to the level of the supersaturation maximum. The additional latent heat release caused by water vapor condensation on large CCN leads to the increase in the vertical velocity, which, in its turn, increases supersaturation (and leads to the increase the level of the supersaturation maximum location). Thus, the decrease in supersaturation caused by condensation of the water vapor condensed on the large CCN is largely compensated by increase in supersaturation caused by increase in the vertical velocity due to corresponding latent heat release. Supplemental experiments show that neglecting latent heat release and decrease in the relative humidity caused by diffusion growth of large CCN below cloud base also leads to an overestimation of the competition effect efficiency.

Results show that the acceleration of the collision rate induced by the largest droplets (grown on large nuclei) is the main factor that accelerates raindrop formation. The role of competition effect seems to be of the secondary importance. This statement requires reconsideration of some cloud seeding hypotheses.

The study has more broad application as concerns the description of the droplet nucleation process in cloud models with detailed microphysics (see review Khain et al., 2000). Most these models do not include aerosol growth, so that aerosol effects on cloud microstructure depend largely on the procedure of droplet nucleation used. The results obtained indicate that the activation of CCN of different size (including large CCN) has a quite small effect on the supersaturation maximum and on droplet concentration.

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References


Segal, Y., Khain, A., in press. Dependence of droplet concentration in different cloud types on aerosol conditions: application to the droplet concentration parameterization. J. Geophys. Res.

