Explicitly Modeling the Effects of Cloud Condensation Nuclei on Warm Rain Initiation

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ABSTRACT: A bin (or spectral) model is developed to investigate the sensitivity of warm rain initiation to cloud condensation nuclei (CCN). It explicitly represents CCN with a formula whose parameters come from the Twomey relationship (or CCN measurements). By seamlessly integrating CCN activation and drop collection with thousands of bins, the model can replicate the effect of CCN on rain initiation, providing a benchmark to test the process parameterizations in rain initiation. The model is used to simulate two extreme cases with CCN parameters of maritime and continental clouds, respectively, where other actual cases usually lie between these two extreme cases. Its simulations show that rain can initiate within half an hour or less as observed in cumulus clouds. The fast rain initiation modeled is attributed mainly to a new process: the condensational conversion of cloud drops to raindrops via collision-coalescence initiators (or drops with radius between 28 and 100 \( \mu m \)). Since the new process is more important in rain initiation than the autoconversion of cloud drops to raindrops when large CCN exist, it is suggested that the process be parameterized into the weather and climate models to better represent CCN and subsequently remove the common bias of “too dense clouds.”

SIGNIFICANCE STATEMENT: The current weather and climate models represent aerosols via implicit parameterizations and have a bias of “too dense clouds.” Their implicit parameterizations of aerosols usually overlook (or misrepresent) some cloud processes. In this paper and its preceding part (Zeng and Li 2020) we proposed a new framework to explicitly parameterize one subset of aerosols: cloud condensation nuclei (CCN). To embody an explicit parameterization of CCN, we still need quantitative information to connect CCN activation and rain initiation, which motivates this study. In the study we developed an accurate microphysical model to simulate the growth of small CCN to large raindrops, providing information on the sensitivity of rain initiation to CCN. We performed many sensitivity simulations and found the condensational conversion of cloud drops to raindrops via collision-coalescence initiators is a vital process in warm rain initiation. Since the process has been overlooked by all the weather and climate models, the study suggests that the process be introduced in the weather and climate models to properly represent the fast warm rain initiation observed and subsequently remove the bias of “too dense clouds.”

KEYWORDS: Aerosols; Cloud microphysics; Condensation; Cloud parameterizations; Parameterization; Aerosol nucleation

1. Introduction

Determining how aerosols affect warm clouds challenges the cloud and climate modeling community (IPCC 2013). In particular, one subset of aerosols, cloud condensation nuclei (CCN), influence cloud drop spectra; in turn, changes to cloud droplet spectra affect rain initiation (Howell 1949; Mordy 1959). However, CCN effect on rain initiation is not represented well in current weather and climate models, bringing about a bias of “too dense clouds” (or the mixing ratio of cloud water is too high) in the models (Nam et al. 2012).

Weather and climate models usually use the one-moment scheme of Kessler (1969) (or its variation) to parameterize warm rain initiation. Why do they still use a simple parameterization developed 50 years ago? The question implies that our knowledge on rain initiation is insufficient, which motivates the present study with two threads: 1) the models are missing an important process, and 2) thus, the parameterization framework of rain initiation is ill-posed.

a. Processes of rain initiation

The classic question of rain initiation come from the radar observations of Squires (1958) and others: “rain can initiate within half an hour or less in cumulus clouds.” Because no rain initiation model with a single process can explain the observations (Blyth and Latham 1993; Blyth et al. 2013), many scientists began to question the classic modeling framework of Mordy (1959) and built complicated models with additional processes or factors, such as giant salt particles (Johnson 1982; Blyth et al. 2003; Jensen and Nugent 2017), radiative effect on microphysics [Roach 1976; Harrington et al. 2000; Zeng 2008, 2018; see Zeng et al. (2022) for review], cloud mixing (Latham and Reed 1977; Telford et al. 1984), and small-scale turbulence (Jonas 1996; Shaw 2003).
While applauding the insight introduced by these complicated models, here we return to the classic modeling framework of Mordy (1959) except for introducing drop collection. After representing CCN activation and drop collection properly with this simple framework (see section 2), the present model replicates the fast rain initiation observed. The fast rain initiation is attributed to the effective cooperation in rain initiation between CCN activation and drop collection, which is studied herein to lay a basis for its parameterization.

b. Parameterization framework

The current parameterization framework of rain initiation cannot accommodate the cooperation between CCN activation and drop collection properly because it usually treats CCN activation and drop collection separately. The parameterization of drop collection in models has evolved over time. The earliest framework for the parameterization, one-moment schemes (Kessler 1969; Cotton 1972), was first proposed where the drops with radius $r > 100\text{ m}$ are classified as raindrops and others as cloud drops. Since the one-moment schemes use only one variable (i.e., cloud water content) and thus have no information on drop number, they cannot represent CCN properly.

Inheriting this framework, many two-moment (or higher-order moment) schemes were developed (Berry and Reinhardt 1974; Tzivion et al. 1987; Khairoutdinov and Kogan 2000; Liu and Daum 2004; Seifert and Beheng 2006; Morrison et al. 2005; Lee and Baik 2017; Zeng and Li 2020). Since the schemes employ two variables (i.e., cloud drop number concentration and cloud water content), they can parameterize CCN explicitly.

To better represent the effect of drop collection on rain initiation, the separation radius between cloud drops and raindrops is decreased from $100\text{ m m}$ (Kessler 1969), to $40\text{ m m}$ (Berry and Reinhardt 1974; Seifert and Beheng 2001; Lee and Baik 2017), and further to $25\text{ m m}$ (Khairoutdinov and Kogan 2000; Zeng and Li 2020). Although the schemes differ in the details of their formulations, most schemes can usually be described by two typical parameterizations, lying at opposite ends of model variability. Figure 1 illustrates these typical parameterizations, where the left-hand ovals represent cloud water, the right-hand ovals represent rainwater, and the red arrows denote processes in rain initiation. In Fig. 1, the autoconversion of cloud water to rainwater represents the increase of rainwater from collision–coalescence growth of cloud droplets. The accretion is the growth of existing rainwater by collecting cloud water (Morrison et al. 2020). The first involves only cloud droplets; the second includes raindrops interacting with cloud droplets. Clearly the definition of the autoconversion and accretion vary with the separation radius.

Collision–coalescence initiators (CCIs), defined as the drops with $28 < r < 100\text{ m m}$, can initiate warm rain effectively (Mason 1971; Johnson 1993; Small and Chuang 2008). The two typical parameterizations in Fig. 1 differ in how they group CCIs. The first classifies CCIs as cloud water, while the second considers CCIs to be rainwater. The first parameterization includes Kessler (1969), and the second follows Khairoutdinov and Kogan (2000) and Zeng and Li (2020) and will be used in this study. Other schemes with separation radius of $40\text{ m m}$ (e.g., Berry and...
The second parameterization in Fig. 1, in contrast to the first one, treats CCIs as rainwater because CCIs can become raindrops quickly via drop collection once they form (Mason 1971; Johnson 1993). Furthermore, its autoconversion (or the collision-induced conversion of cloud drops to CCIs) can be parameterized accurately (Zeng and Li 2020) and its condensational growth of cloud droplets to CCIs be parameterized explicitly. In addition, the second parameterization can express the sedimentation of rainwater properly in terms of the average radius of raindrops (including CCIs). For example, if an air parcel contains only CCIs, its rainwater sediments with the small terminal velocity of CCIs.

The second parameterization illustrated in Fig. 1 will be studied in this paper. In contrast to other two-moment schemes (e.g., Seifert and Beheng 2001; Khairoutdinov and Kogan 2000; Lee and Baik 2017), the scheme introduces a new process, the condensational conversion of cloud water to rainwater via CCIs. The new process is independent of the autoconversion of cloud water to rainwater. It consists of two steps: cloud droplets grow to CCIs via condensation first and then CCIs grow to raindrops quickly by accreting cloud drops. To test the new process, a bin model is developed that includes drop collection and thus can simulate the growth of small CCN to large raindrops. The model and its simulations are introduced in this paper with six sections. The model structure is introduced in section 2 with a CCN representation. Simulations are presented in sections 3 and 4 to show 1) the model replicates the Twomey relationship and 2) rain initiation is sensitive to CCN, respectively. Section 5 outlines how to apply the simulation results to rain initiation parameterization, and section 6 summarizes.

2. Model description

A bin model is developed based on Mordy (1959) and Zeng (2018) to explicitly simulate CCN activation and drop collection. It is introduced in this section with numerical schemes, CCN representation, and model structure.

a. Representing drop collection and condensation growth in the Lagrangian system

The model adopts the framework of the bin model of Zeng (2018) that is based on the method of Kovetz and Olund (1969). Specifically, it simulates drop growth in the Lagrangian system by tracking drops with thousands of bins. Let $m_j$ denote drop mass of a bin. Then $m_j$ is increased (or decreased) to represent the condensation growth (or evaporation shrinkage) of drops, avoiding the spurious spectrum broadening that often happens in the bin models in the Eulerian system (Khain et al. 2008, 2010, 2012; Flossmann and Wobrock 2010; Iguchi et al. 2012).

The model, following Kovetz and Olund (1969), simulates drop collection with mass conservation. Consider drops with number $N'$ and mass $m'$ that are newly formed in a collision process. When $m_j \leq m' < m_{j+1}$, the model interprets the drops as $N'_j$ drops at bin $j$ and $N'_{j+1}$ drops at bin $j + 1$, based on the conservation of drop number and mass or

$$N'_j + N'_{j+1} = N', \quad (1a)$$

$$m'_j N'_j + m'_{j+1} N'_{j+1} = m' N', \quad (1b)$$

which is read as

$$N'_j = \frac{m'_{j+1} - m'}{m'_{j+1} - m_j} N', \quad (2a)$$

$$N'_{j+1} = m'_{j+1} - m_j N'. \quad (2b)$$

Although the method of Kovetz and Olund [or Eq. (1)] is reasonable in physics, it received unfair comments for a long time. Since the method with ~40 bins generates too many large drops (Scott and Levin 1975), this error of numerical spectrum broadening was attributed to the method (Khain et al. 2000). In fact, the error origins in the coarse bin resolution near drop radius 40 μm because of the great variation of the collection kernel there. After increasing the bin number to 1024, the method outputs accurate results in comparison to the analytic solution of the stochastic collection equation (SCE; Zeng 2018).

The present model is like the model of Pinsky and Khain (2002), to some extent, for both the models use thousands of bins to explicitly simulate CCN activation. On the other hand, the models differ in CCN representation because the present model incorporates the effects of not only CCN size but also composition via the Twomey relationship (see section 2b).

In addition, the two models differ in drop collection simulation [or Eq. (2)]. Pinsky and Khain (2002) used the two-step method of Bott (1998) to simulate drop collection. In the first step drops with mass $m'$ newly formed in a collision process is added to bin $j$ if $m_j \leq m' < m_{j+1}$. In the second step a fraction of drops is transported from bin $j$ to $j + 1$, where the fraction depends on an imposed pattern of drop spectrum between $m_j$ and $m_{j+1}$. The pattern (or fraction) is adjusted so that the modeled drop spectrum fits the analytic solution of SCE in specific cases (e.g., those with the Golovin collection kernel; Bott 1998), which is referred to as “hard adjustment.” In the method the “hard adjustment” satisfies (1b) but violates (1a).

The “hard adjustment” was often used in the models with bin number ~ 40 (Berry and Reinhardt 1974; Zeng and Zhang 1989; Bott 1998). Since the “hard adjustment” was tested only in some specific cases, whether it works in other cases (e.g., those with condensational growth) is still unknown. In contrast, the method of Kovetz and Olund [or Eq. (1)] is general and consequently can work in any cases given sufficient bin resolution. Hence the present model employs the method of Kovetz and Olund with 4096 bins.

b. CCN representation

The model simulates CCN activation explicitly just as the other Lagrangian models (Mordy 1959; Pinsky and Khain 1974; Seifert and Beheng 2001; Lee and Baik 2017).
Different from the other models, it uses a spectrum expression of condensation nuclei (CN) so that it can replicate the Twomey relationship between active CCN and supersaturation (Twomey 1959).

The spectrum expression of CN is derived as follows, beginning with the observed Twomey relationship that usually exhibits a power-law dependence of active-CCN number concentration $N_{CCN}$ on supersaturation $S_{max} - 1$ (Twomey 1959), or

$$N_{CCN} = N_{CCN1} [100(S_{max} - 1)]^k,$$

where $k$ is a constant and $N_{CCN1}$ is the number concentration of active CCN at $S_{max} - 1 = 1\%$. The saturation ratio $S_{max}$ records the historical maximum of the instantaneous saturation ratio $S_n$ in an air parcel. In other words, it corresponds to $m_{min}$ the minimum salt mass of activated CN or the maximum salt mass of unactivated CN. The theory of CCN activation connects $S_{max}$ and $m_{min}$ by (Twomey 1959; Rogers and Yau 1989, p. 89)

$$S_{max} - 1 = \left( \frac{4C_1^3}{27C_2 m_{min}} \right)^{1/2},$$

where $C_1$ and $C_2$ are two coefficients of the drop growth equation (see section 2c).

Let $N_{CN}(m)$ denote the number concentration of CN with salt mass less than $m$. Thus $N_{CN}(m_{min})$ denotes the number concentration of unactivated CN. Hence, $N_{CN}(m_{min}) + N_{CCN}(m_{min})$ represents the total number concentration of unactivated and activated CN. Since CCN activation does not change the total number concentration of CN, $d[N_{CN}(m_{min}) + N_{CCN}(m_{min})] = 0$, yielding $dN_{CN}(m_{min}) = -dN_{CCN}(m_{min})$. Substituting (4) into (3) and then differentiating the resulting equation with respect to $m_{min}$ gives a new expression of CCN spectrum. That is,

$$\frac{dN_{CN}(m_{min})}{d \ln m_{min}} = -\frac{dN_{CCN}(m_{min})}{d \ln m_{min}} = 100^k N_{CCN1} \frac{k}{2} \frac{4C_1^3}{27C_2} m_{min}^{-k/2},$$

which is rewritten as

$$\frac{dN_{CN}(m)}{d \ln m} = 100^k N_{CCN1} \frac{k}{2} \frac{4C_1^3}{27C_2} m^{-k/2}.$$

If all aerosols are CN and $k = 2$, (6) degenerates into the Junge distribution (Jiusto and Lala 1981; Rogers and Yau 1989).

The bin model uses (6) to represent CN. Its two parameters—$k$ and $N_{CCN1}$—are available from the CCN observations (Twomey 1959; Braham 1976; Hudson 1993). In this paper, two extreme cases are discussed: $k = 0.4$ and $N_{CCN1} = 100$ cm$^{-3}$ for maritime clouds, and $k = 0.9$ and $N_{CCN1} = 500$ cm$^{-3}$ for continental clouds. Other actual cases usually lie between these two extreme cases (Jiusto 1967).

Given $k$ and $N_{CCN1}$, (6) still needs the information of CN composition. For the sake of simplicity, CN are assumed to be composed of sea salt (NaCl). In fact, the present CN representation is suitable for other salt or multicomposition particles after replacing the particles with their equivalent CN of NaCl, where the equivalent CN of NaCl are defined as the virtual NaCl particles whose Twomey relationship equals that of real aerosol particles.

In summary, (6) is equivalent to the Twomey relationship if all drops are close to equilibrium with water vapor, which is illustrated in Fig. 2. Since the bin model uses (6) as an input, it
generates the Twomey relationship as an output (when its initial drops are at equilibrium with water vapor), which supports the equivalence between (6) and the Twomey relationship (see section 3 for simulation details or the circles in Fig. 2 for simulation results).

On the other hand, drops with large CCN in real clouds are rarely at equilibrium with water vapor because they reach the equilibrium in hours to days which depends on the mass of large CCN (Kogan 1991). Hence the Twomey relationship represents large CCN in real clouds inaccurately. If a model used the Twomey relationship as an input to represent CCN in clouds (Khain et al. 2008), it introduced a representation error of large CCN.

Different from the Twomey relationship, (6) can be used to represent CCN in real clouds. It is connected to the Twomey relationship as follows. Suppose we sample aerosols in a cloud and then move the sample of the aerosols into a cloud chamber for CCN measurement. Since the aerosols are at equilibrium with water vapor in the cloud chamber, they satisfy the Twomey relationship and thus their aerosols for CCN measurement. Since the aerosols are at equilibrium with water vapor in the cloud chamber, they satisfy the Twomey relationship and thus their aerosols are obtained. After obtaining $k$ and $N_{CCN}$, we assume that the aerosols besides the sampled ones satisfy (6) with the same $k$ and $N_{CCN}$, no matter whether they satisfy the Twomey relationship in real clouds.

c. Air parcel model

The bin model is used to simulate an air parcel that moves upward with vertical velocity $w$. The parcel contains CN and drops. Its saturation ratio is governed by

$$
\frac{dS_w}{dt} = Q_{w1}(T)w - Q_{w2}(T)\sum \frac{dm_j}{dt},
$$

where $S_w$ is the saturation ratio with respect to liquid water, $T$ temperature, and $m_j$ ($j = 1, 2, \ldots$) the mass of all drops with different sizes. The thermodynamic variables

$$
Q_{w1}(T) = \frac{1}{T} \left( \frac{L_v g}{R_y C_p T} - \frac{g}{R_y} \right)
$$

and

$$
Q_{w2}(T) = \rho_a \left( \frac{R_T}{E_w(T)} + \frac{R \ln L_y^2}{\rho R_y C_p T} \right),
$$

where $p$ is air pressure, $g$ the acceleration due to gravity, $L_v$ the latent heat of vaporization, $C_p$ the specific heat of dry air, $R_y/R$, the gas constant for dry air/water vapor, $E_w$ the saturation vapor pressure over water, and $\rho_a$ air density. The diffusional drop growth rate

$$
\frac{dm}{dt} = \frac{4\pi r}{A_w + B_w} \left( S_w - 1 - C_1 \frac{r}{r^3} + C_2 \frac{m_j}{r^3} \right),
$$

where $r$ is drop radius, $m_j$ the mass of the dissolved NaCl,

$$
A_w = \frac{L_v}{f_{d} Q K T} \left( \frac{L_v}{R_T} - 1 \right),
$$

$$
B_w = \frac{R_T}{f_{d} K T},
$$

$$
C_1 = \frac{2\sigma_{vd}}{\rho R_T T},
$$

$$
C_2 = \frac{3\sigma_{vd} M_w}{4\pi \rho R_T T},
$$

where $D$ is the diffusivity of water vapor, $K$ the coefficient of thermal conductivity of dry air, $\sigma_{vd}$ the surface tension of liquid–vapor interface, $l_{vd}$ the van’t Hoff factor, $\rho$ the density of liquid water, $f_m/f_Q$ the ventilation factor for mass transfer/thermal diffusion, $M_w/M_s$ the molecular weight of dissolved salt/water, and $F_{d}/F_{d}$ the factor for the kinetic effect in heat/water vapor diffusion.

The bin model is used to simulate an air parcel at an altitude of pressure $p = 800$ hPa and temperature $T = 15^\circ C$. It uses (6) to represent CN spectrum and sets the maximum salt mass $m_{max} = 10^{-11} g$ except for specification.

Suppose that all drops stay at equilibrium with water vapor initially. Given an initial saturation ratio of $S_w$, CN arise as solution drops with different size. Equation (10) shows that the radius $r$ of a solution drop is related to its salt mass $m_j$ by

$$
S_w - 1 - C_1 r + C_2 m_j = 0.
$$

Two initial drop spectra are used in sections 3 and 4, respectively. In section 3, all drops are assumed to be at equilibrium at $S_w = 1$ initially. They are discretized with 4096 bins, denoted with drop mass $m_j$ ($j = 1, 2, \ldots, 4096$). One-half of the bins are used to represent solution drops whose salt mass ranges from $10^{-18} g$ to $m_{max}$ (e.g., $10^{-11} g$). To be specific, the logarithm of salt mass is discretized uniformly between $\ln 10^{-18} g$ and $\ln m_{max}$ with 2048 bins. As a result, the initial drop mass in each bin is determined by (12), and subsequently $m_{2048}$ corresponds to a salt mass of $m_{max}$.

The other half of the bins are reserved to represent large drops that will form via drop collection, which are denoted as

$$
m_j = m_{2048} \exp[(j - 2048/a)],
$$

where $j > 2048$ and $a = 363.6$. To maintain the continuity in salt mass between bins, all the bins with $j > 2048$ are assumed to take the same salt mass per drop as bin 2048 (or $m_{max}$).

Since there are no drops at $j > 2048$ initially, the initial number concentration of drops at $j > 2048$ is set to be zero. Once the drop mass in all the bins (i.e., $m_j, j = 1, 2, \ldots, 4096$) are set initially, the same algorithm is used to simulate both drop condensation and collision in all the bins. Thus, there is no difference in algorithm between the bins with $j \leq 2048$ and $j > 2048$.

Equations (7) and (10) are integrated numerically, following Zeng (2018). For simplicity, $T$ and $p$ are assumed to be constant. In addition, (10) is integrated implicitly with a small time step of 0.05 s so that CCN activation is simulated accurately (see appendix).
3. Replication of the Twomey relationship

The Twomey relationship was often misused to represent CCN in cloud modeling by overlooking its condition (or the environment for drops in cloud chamber) (e.g., Khain et al. 2008, 2010, 2012; Iguchi et al. 2012), where the condition involves the equilibrium between drops and water vapor and thus does not exist in all clouds. To avoid the misuse of the Twomey relationship, the present model employs (6) to represent CN instead.

A result, the models satisfy the Twomey relationship when its initial drops are at equilibrium with water vapor, which is tested in this section via bin model simulations.

Experiment M1I is designed to simulate a marine cloud with $w = 1 \text{ m s}^{-1}$, $p = 800 \text{ hPa}$, and $T = 15^\circ\text{C}$. It chooses $k = 0.4$, $N_{\text{CCN}} = 100 \text{ cm}^{-2}$, and $m_{\text{max}} = 10^{-11} \text{ g}$ to represent marine CCN. It incorporates vapor condensation but no drop collection. Its modeled drop spectrum is displayed in Fig. 3, showing CCIs but no raindrops form. For comparison, another experiment of C1I is carried out to obtain the sensitivity of CCIs to CCN. It takes the same setup as M1I except for continental CCN. It uses $k = 0.9$, $N_{\text{CCN}} = 500 \text{ cm}^{-2}$ and $m_{\text{max}} = 10^{-11} \text{ g}$ to represent continental CCN. Its modeled drop spectrum resembles that of M1I except for the water content of CCIs (figure omitted).

Since the two experiments incorporate no drop collection, their CCIs form via the condensation growth of cloud drops. To be specific, since their relative humidity $S_w > 1$, water vapor deposits on drops, bringing about an increase of liquid water content (LWC; see Fig. 4). As a result, a part of cloud drops grows to CCIs via condensation.

The modeled number concentration of activated CCN (or the number concentration of cloud drops and CCIs) satisfies the Twomey relationship. Using the same setup as M1I and C1I, five additional sets of experiments are carried out that take a vertical velocity of 0.1, 0.2, 0.5, 3, or 10 m s$^{-1}$, respectively. Their maximum number concentrations of activated CCN are displayed against the maximum supersaturation by circles in Fig. 2. Clearly, the modeled concentration of activated CCN is close to the Twomey relationship, supporting the equivalence between (6) and the Twomey relationship.

4. Sensitivity of rain initiation to CCN

Since M1I was designed to test whether the model satisfies the Twomey relationship, its initial drops were assumed to be at equilibrium at $S_w = 1$. However, large drops need hours or days to reach their equilibrium radius at $S_w = 1$ (Kogan 1991).
Table 1. Parameters of the control experiments. Experiment naming convention starts with a letter M or C for a marine or continental cloud, followed by a number of vertical velocity, and ends with a letter I for special initial drop spectrum or C for an experiment with drop collection.

<table>
<thead>
<tr>
<th>Expt</th>
<th>w (m s⁻¹)</th>
<th>K</th>
<th>$N_{CCN1}$ (cm⁻³)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>M1I/C1I</td>
<td>1</td>
<td>0.4/0.9</td>
<td>100/500</td>
<td>Testing the Twomey relationship when initial $S_a = 1$ (or Fig. 2); also for sensitivity of CCI initiation to initial drop spectrum</td>
</tr>
<tr>
<td>M3/C3</td>
<td>3</td>
<td>0.4/0.9</td>
<td>100/500</td>
<td>Default experiments with no drop collection; for sensitivity of CCI initiation to CCN (or Fig. 7)</td>
</tr>
<tr>
<td>C3C</td>
<td>3</td>
<td>0.9</td>
<td>500</td>
<td>High vertical velocity with drop collection; for sensitivity of rain initiation to CCN at high w (or Fig. 9)</td>
</tr>
<tr>
<td>M01C</td>
<td>0.1</td>
<td>0.4</td>
<td>100</td>
<td>Low vertical velocity with drop collection; for sensitivity of rain initiation to CCN at low w (or Fig. 10)</td>
</tr>
</tbody>
</table>

Hence, M1I implicitly introduced spurious large initial droplets if it addressed a real cloud. To avoid the spurious initial large droplets, experiments with a new initial drop spectrum are carried out in this section (see Table 1 for experiment summary), focusing on how CCN impact the initiation of CCI and raindrops given the air vertical velocity $w$ and the maximum salt mass $m_{\text{max}}$.

a. Condensational formation of CCIs

A default experiment, M3, is designed to test the condensational formation of CCIs. It takes the same setup as M1I except for $w = 3$ m s⁻¹ and a new initial drop spectrum. Its initial drop spectrum is similar to that of Mordy (1959).

Mordy (1959) assumed that at cloud base drops formed on nuclei smaller than 0.12 $\mu$m are at equilibrium at 100% relative humidity, while the drops formed on nuclei larger than 0.12 $\mu$m are at equilibrium at 90% relative humidity. However, the discontinuity of relative humidity at nuclei 0.12 $\mu$m brings about an abnormal phenomenon: some drops formed on large nuclei are smaller than those formed on small nuclei. To avoid the abnormal phenomenon, the relative humidity for drop equilibrium is assumed to decrease linearly with nuclei mass from 100% at $m_i = 0$% to 90% at $m_i = 2.5 \times 10^{-14}$ g (or dry sea-salt particle radius 0.14 $\mu$m) and maintain 90% at $m_i > 2.5 \times 10^{-14}$ g. The drop spectrum at cloud base is then obtained with (6) and (12).

Experiment M3 uses the preceding drop spectrum at cloud base as initial drop spectrum. Like C1I, it uses 4096 bins to discretize the drop spectrum, one-half of which are used to represent solution drops whose salt mass ranges from $10^{-15}$ g to $m_{\text{max}}$ (i.e., $10^{-11}$ g) and the other half are reserved to represent large drops that will form via drop collection with $a = 246.2$. Its modeled drop spectrum is displayed in Fig. 5. At 30 min, most of the drops are located with radius between 28.5 and 31.2 $\mu$m, showing that condensation does bring about CCIs even after the new initial drop spectrum is used.

For comparison, another default experiment of C3 is carried out to obtain the sensitivity of CCIs to CCN. C3 takes the same setup as M3 except for continental CCN (i.e., $k = 0.9$, $N_{CCN1} = 500$ cm⁻³, and $m_{\text{max}} = 10^{-3}$ g). Its CCIs, as shown in Fig. 6, form more slowly than the counterpart of M3. If CCI initiation is defined as the water content of CCIs above $10^{-3}$ g m⁻³, CCIs initiate at 21.1 and 68.7 min in M3 and C3, respectively.

To study the condensational formation of CCIs extensively, $3 \times 9 \times 2$ experiments are carried out that take the same setup as M3 and C3 except for $w$ and $m_{\text{max}}$. They use the following parameters: $w = 1, 3, 5$ m s⁻¹; $m_{\text{max}} = 10^{-13}, 3 \times 10^{-13}, 10^{-12}, 3 \times 10^{-12}, 10^{-11}, 3 \times 10^{-11}, 10^{-10}, 3 \times 10^{-10},$ or $10^{-9}$ g; $k = 0.4/N_{CCN1} = 100$ cm⁻³, or $k = 0.9/N_{CCN1} = 500$ cm⁻³. Their modeled time of CCI initiation is displayed in Fig. 7, showing that the CCI initiation is sensitive to CCN and $w$. To be specific, given $w$ and $m_{\text{max}}$, the difference in CCI initiation time between the two panels (or the difference between two lines with the same color) indicates that the marine clouds with

![Fig. 5](image1.png)  
**Fig. 5.** As in Fig. 3, but for Exp. M3 where initial large drops are not at equilibrium with water vapor.

![Fig. 6](image2.png)  
**Fig. 6.** Time series of CCI water content in Exp. M3 with marine CCN (blue) and Exp. C3 with continental CCN (red).
$k = 0.4/N_{CCN1} = 100 \text{ cm}^{-3}$ initiate CCIs faster than their continental counterparts with $k = 0.9/N_{CCN1} = 500 \text{ cm}^{-3}$. In addition, each line in a panel indicates that a cloud, given $w$, initiates CCIs faster with increasing $m_{\text{max}}$. 

b. Collision-induced conversion of cloud drops to raindrops

Once CCIs form via condensation growth, they grow to raindrops quickly via drop collection. Thus, the condensational conversion of cloud drops to raindrops via CCIs is independent of the collision-induced conversion of cloud drops to raindrops, which is discussed in this subsection.

Experiment C3C, in contrast to C3, is designed to show the formation of raindrops by drop collection. C3C takes the same setup as C3 except for incorporating drop collection, where the collection kernel is computed with the drop terminal velocity of Beard (1976) and the collection efficiency of De Almeida (1977) complemented by Mason (1971).

Its modeled drop spectrum is displayed in Fig. 8, showing that raindrops form in $\sim 15$ min. Since the condensational formation of CCIs is weak during the rain initiation (as shown in Fig. 6), the CCIs that grow to raindrops originate in the self-collection of cloud drops, indicating that the collision-induced conversion of cloud drops to raindrops is more important than the condensational conversion in the experiment.

c. Sensitivity to CCN at high vertical velocity

To study the sensitivity of rain initiation to CCN and $w$, another $3 \times 9 \times 2$ experiments are carried out that take the same setup as C3C except for $w$ and CCN. Their parameters are the same as those in Fig. 7. That is, $w = 1, 3,$ or $5 \text{ m s}^{-1}$; $m_{\text{max}} = 10^{-12}, 3 \times 10^{-13}, 10^{-12}, 3 \times 10^{-12}, 10^{-11}, 3 \times 10^{-11}, 10^{-10}, 3 \times 10^{-10},$ or $10^{-9}$ g; $k = 0.4/N_{CCN1} = 100 \text{ cm}^{-3}$, or $k = 0.9/N_{CCN1} = 500 \text{ cm}^{-3}$. Their modeled time of rain initiation is displayed in Fig. 9, where rain initiation is defined as the rainwater content (or the water content of raindrops with $r > 100 \mu m$) above $10^{-3} \text{ g m}^{-3}$.

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1 The rapid fluctuation of drop spectrum near $25 \mu m$ at 20 min in Fig. 8 comes from the discontinuous growth of drops as well as the discontinuity of initial CN spectrum at $m_s = m_{\text{max}}$. A drop with mass 10, for example, collects another drop with mass 9, forming a new drop with mass 19. As a result, its mass jumps from 10 to 19, manifesting a discontinuous increase in drop mass. The discontinuous drop growth brings about the rapid fluctuation near $25 \mu m$ via the discontinuity of initial drop spectrum at $m_s = m_{\text{max}}$ or bin 2048 that is caused by the observed discontinuity of CN spectrum at $m_s = m_{\text{max}}$ (Woodcock 1953; Jiusto 1967).
Figure 9 shows that rain initiates earlier with increasing $w$ and $m_{\text{max}}$. In addition, rain with marine CCN initiates earlier than that with continental CCN. In general, rain can initiate within half an hour or less, which agrees with the radar observations of rain initiation in cumulus clouds (Squires 1958; Saunders 1965).

The sensitivity of rain initiation to CCN is attributed mainly to the sensitivity of CCI initiation to CCN because CCIs collect cloud drops and subsequently become raindrops quickly. The sensitivity is explained by comparing two kinds of CCI formations: the condensational conversion and the collision-induced conversion of cloud drops to CCIs. In the experiments in Fig. 9, the condensational conversion is more sensitive to CCN than the collision-induced conversion, although they are mixed. To separate the two conversions, the experiments in Fig. 7 are revisited that exclude drop collection and consequently possess only the condensational conversion. Owing to the similarity in the experiments between Figs. 7 and 9 except for drop collection, the condensational conversion in the experiments in Fig. 7 can be substituted for its counterpart in the experiments in Fig. 9 approximately.

Suppose that the CCI initiation time in Fig. 7 represents the condensational CCI formation in Fig. 9. Thus, the (positive) difference between the CCI initiation time in Fig. 7 (or the dashed lines in Fig. 9) and the rain initiation time in Fig. 9 can be used to infer the collision-induced CCI formation in the experiments in Fig. 9. When the CCI initiation time in Fig. 7 is much shorter (or longer) than the rain initiation time in Fig. 9, CCIs form mainly by the condensational growth of cloud drops (or the collection between cloud drops). Hence the difference between the CCI initiation time (dashed lines) and the raindrop initiation time (solid lines) in Fig. 9 indicates that the condensational conversion of cloud drops to CCIs is more important than the collision-induced conversion when the maximum salt mass $m_{\text{max}} > 1.5 \times 10^{-10} \text{g}$ (or dry sea-salt particle radius $>2.5 \mu\text{m}$).

In Fig. 9, the rain initiation is not as sensitive to CCN as the condensation-induced CCI initiation because of coexistence of the collision-induced conversion and the condensational conversion of cloud drops to CCIs. Since the collision-induced conversion “dilutes” the condensational conversion, the sensitivity of rain initiation to CCN is reduced, which is understood from two sides. On one side, condensation not only brings about CCIs but also accelerates the collision-induced conversion via collision efficiency. The collision efficiency between two drops increases markedly as the size of the collector drop (with radius $>20 \mu\text{m}$) increases (Mason 1971; De Almeida 1977). Since condensation increases the size of drops and thus the collision efficiency between drops, condensation accelerates the collision-induced conversion via collision efficiency and subsequently contributes to the sensitivity of rain initiation to CCN. On the other side, the condensational CCI formation is more sensitive to CCN than the collision-induced CCI formation, which is understood from the similarity between the collection kernel and time in the Eulerian system of SCE (Srivastava 1988). When the collection kernel is doubled, for example, the time for rain initiation is halved. Since the collection kernel is proportional to the collision efficiency, an increase in collision efficiency, caused by the condensation-induced shifting of drop spectrum peak toward raindrops (see Fig. 5), can accelerate the accretion of cloud droplets by relatively large droplets and CCIs and subsequently accelerate rain initiation.
other side, the collision-induced conversion is proportional to the square of LWC (Srivastava 1971; Zeng and Li 2020) and LWC increases mainly with \( w \). As a result, high \( w \) brings about strong collision-induced conversion so that the sensitivity of rain initiation to CCN looks small relative to the sensitivity of condensation-induced CCI initiation to CCN.

d. Sensitivity to CCN at low vertical velocity

To reveal the sensitivity of rain initiation to CCN at low \( w \), 9 \( \times \) 2 experiments are carried out that take the same setup as C3C except for \( w = 0.1 \text{ m s}^{-1} \) and CCN. They use the same CCN parameters as those in Fig. 9 except with different vertical velocity. Their modeled time of rain initiation is displayed in Fig. 10, show that rain initiation is quite sensitive to CCN.

To show whether the condensational conversion plays an important role in rain initiation, another 9 \( \times \) 2 experiments are carried out that take the same setup as C3C and the other experiments in Fig. 10 except for no drop collection. Their time of CCI initiation is displayed by the dashed lines in Fig. 10, approximately representing the time of condensation-induced CCI initiation in the counterparts with drop collection. The difference in time between dashed and solid lines with the same color in Fig. 10 indicates that the condensational conversion is more important than the collision-induced conversion when the maximum salt mass \( m_{\text{max}} > 4 \times 10^{-11} \text{ g} \) (or dry sea-salt particle radius \( > 1.4 \mu \text{m} \)).

At \( m_{\text{max}} < 4 \times 10^{-11} \text{ g} \), the condensational conversion still contributes to rain initiation and consequently brings about the high sensitivity of rain initiation to CCN. To be specific, the condensation-induced shifting of cloud drop spectrum peak toward raindrops (see Fig. 5) accelerates the collision-induced conversion via collision efficiency (see footnote 2 for explanation). This effect of condensation on rain initiation is relatively important at low \( w \) because low \( w \) brings about low LWC that in turn brings about relatively low collision-induced conversion. In addition, the effect is embodied via another form of the condensational conversion of cloud drops to CCIs: two small cloud drops collide to form a large new cloud drop that in turn grows to a CCI via condensation. Obviously, this form of the condensational conversion is different from the form that a cloud drop grows directly to a CCI via condensation. Although this form of the condensational conversion is not represented by the dashed lines in Fig. 10, it does contribute to the sensitivity of rain initiation to CCN at \( m_{\text{c}} < 4 \times 10^{-11} \text{ g} \) in Fig. 10, explaining the sensitivity of rain initiation to \( m_{\text{max}} \) at \( m_{\text{max}} < 4 \times 10^{-11} \text{ g} \).

5. Discussion

The modeled sensitivities of rain initiation to CCN provide clue to parameterize warm rain initiation. After CCIs are grouped as a part of rainwater (Fig. 1), the bulk conversion of cloud water to rainwater is expressed in terms of three parameterizations: the autoconversion, the accretion, and the condensational conversion of cloud water to rainwater (see the three red arrows between cloud water and rainwater in Fig. 1). Since the accretion works only after rain initiation, the autoconversion and condensational conversion during rain initiation are compared with the aid of Figs. 9 and 10. Roughly speaking, the dashed lines in Figs. 9 and 10 provide information on the condensational conversion in Fig. 1; the difference in time between the dashed and solid lines provides information on the autoconversion. The two conversions, as shown in Figs. 9 and 10, have different importance in different cases. The autoconversion and condensational conversion are dominant when the maximum salt mass \( m_{\text{max}} \) are small and large, respectively.

The separation between the small and large \( m_{\text{max}} \) depends on vertical velocity and the drop spectrum at cloud base (or initial drop spectrum). The separation shifts to lower \( m_{\text{max}} \) with decreasing \( w \) (see Figs. 9 and 10). It also shifts to lower \( m_{\text{max}} \) with increasing the relative humidity (RH) of the equilibrium of initial large drops (see the difference in initial drop spectrum between Figs. 3 and 5 whose initial large drops are at equilibrium with water vapor RH = 100% and 90%, respectively; the initial drop spectrum in Fig. 5 is too low to be displayed and thus only the drop spectrum at 1 min is displayed instead). Specifically, when initial large drops are at equilibrium with water vapor at RH = 90% (see section 4), the small and large \( m_{\text{max}} \) is separated at salt particle mass \( \sim 1.5 \times 10^{-10} \text{ g} \) (or dry sea-salt particle radius \( \sim 2.5 \mu \text{m} \)) with vertical velocity around 1 m s\(^{-1}\) and salt particle mass \( \sim 4 \times 10^{-11} \text{ g} \) (or dry sea-salt particle radius \( \sim 1.4 \mu \text{m} \)) with a vertical velocity of 0.1 m s\(^{-1}\).
In contrast, when initial large drops are at equilibrium with water vapor at RH = 100% (see section 3), CCIs are initiated at cloud base (see Fig. 3) and consequently the small and large \(m_{\text{max}}\) is separated at salt particle mass \(< 6 \times 10^{-12} \text{ g}\) (or dry sea-salt particle radius \(< 0.86 \mu \text{m}\)), because a drop with radius 28 \(\mu\text{m}\) and salt mass \(6 \times 10^{-12} \text{ g}\) is at equilibrium with water vapor at RH = 100%. Roughly speaking, the small and large \(m_{\text{max}}\) is separated at salt particle radius 0.5 \(\mu\text{m}\), which is estimated after taking account of another process: two small cloud droplets collide to form a large new droplet that in turn grows to a CCI via condensation (please note that the process contributes to the solid lines but not the dashed lines in Figs. 9 and 10).

Since the two conversions of cloud water to rainwater function quite differently, it is imperative to distinguish them. First, the autoconversion is proportional to the square of LWC and is also affected by CCN via CCN activation or the number concentration of cloud drops with a given LWC (Srivastava 1971; Zeng and Li 2020). Second, the condensational conversion depends on other variables. To be specific, the condensational conversion rate of drop number from cloud drops to CCIs is expressed as

\[
\frac{\partial N(m, t)}{\partial m} \bigg|_{m = m^*} \frac{dm}{\partial t} \bigg|_{m = m^*},
\]

where \(N(m, t)\) denotes the number concentration of drops with drop mass less than \(m\) at time \(t\), \(m^*\) is the mass of a drop with radius 28 \(\mu\text{m}\), and \(dm/\partial t\) is given in (10). Since \(\partial N(m, t)/\partial m\) \(|_{m = m^*}\) is affected by both drop collection and CCN spectrum, the condensational conversion is affected by CCN spectrum, too, especially by \(m_{\text{max}}\).

When \(m_{\text{max}}\) is large, the condensational conversion includes the contribution of giant CCN (GCCN), where the contribution is embodied via a series of processes: GCCN grow via condensation to CCIs that in turn collect cloud drops to become raindrops. Obviously, the condensational conversion is much more effective than the process that GCCN grow directly to raindrops via condensation (Jensen and Nugent 2017).

The sensitivity of rain initiation to the maximum salt mass \(m_{\text{max}}\) in Figs. 9 and 10 makes sense. Consider the tail of drop spectrum at radius \(> 28 \mu\text{m}\) and at 20 min in Fig. 8. Since the vertical axis of Fig. 8 is scaled logarithmically, the tail is small. However, it is the small tail that leads to rain initiation. If additional salt particles with mass \(< m_{\text{max}}\) and low number concentration were introduced, the tail would become large and thus rain be initiated faster. In short, the tail of drop spectrum for rain initiation is affected by the tail of salt particle spectrum at mass \(< m_{\text{max}}\).

The maximum salt mass \(m_{\text{max}}\) corresponds to the minimum \(S_v\) for CCN activation in the Twomey relationship. When \(S_v\) is below the minimum \(S_v\), no drops grow from CCN in a cloud chamber, indicating no salt particles with mass \(> m_{\text{max}}\). The CCN observations show that both the minimum \(S_v\) and \(m_{\text{max}}\) vary greatly from case to case (Woodcock 1953; Jiusto 1967).

The introduction of \(m_{\text{max}}\) does not violate the Twomey relationship. When \(m_{\text{max}}\) is large, (6) can be used to describe the existence of GCCN (Woodcock 1953; Jensen and Nugent 2017), small, nonexistence of GCCN. Since \(m_{\text{max}}\) is an independent variable to describe large CCN in the framework of the Twomey relationship, it can be introduced into the process parameterizations to better represent the effect of CCN on rain initiation.

6. Conclusions

A bin model is developed to accurately simulate the sensitivities of rain initiation to CCN. It uses Eq. (6) to represent CCN whose parameters come from the observed Twomey relationship or CCN measurements. It also uses thousands of bins to seamlessly integrate CCN activation and drop collection, providing a benchmark to test the process parameterizations in rain initiation.

The model is used to simulate two extreme cases with CCN parameters of maritime and continental clouds (i.e., \(k = 0.4/N_{\text{CCN1}} = 100 \text{ cm}^{-3}\), and \(k = 0.9/N_{\text{CCN1}} = 500 \text{ cm}^{-3}\)), respectively. Since other actual cases usually lie between these two extreme cases (Jiusto 1967), it is inferred that other actual cases possess rain initiation time between those of the two extreme cases. Hence, the modeling results of the two extreme cases provide climatological information on rain initiation.

The model results (or Fig. 9) show that rain can initiate within half an hour or less as observed in cumulus clouds (Squires 1958; Saunders 1965). They (or Fig. 10) also show that rain initiation is sensitive to CCN. This fast rain initiation modeled and the sensitivity of rain initiation to CCN come from the effective cooperation between CCN activation and drop collection, and the effective cooperation is embodied mainly by a new process: the condensational conversion of cloud drops to raindrops via CCIs.

The condensational conversion of cloud drops to raindrops via CCIs is independent of the autoconversion of cloud water to rainwater (see Fig. 1). It consists of two steps: cloud drops grow to CCIs by condensation and then CCIs grow quickly to raindrops by accreting cloud drops. The condensational conversion and autoconversion, as shown by the bin model simulations, have different importance in different cases. Roughly speaking, the condensational conversion is more important than the autoconversion when large CCN exists (or the dry salt particle radius of the largest CCN with mass \(m_{\text{max}} > 0.5 \mu\text{m}\)); otherwise, the autoconversion is more important (see Figs. 6 and 8).

Since the condensational conversion has not been represented by the current parameterization schemes, its parameterization will be developed based on the bin model simulations. Theoretically, the parameterization can benefit the weather and climate models for better representing CCN especially large CCN. To be specific, the current weather and climate models do not represent clouds well. They have a bias of “too dense clouds” (Nam et al. 2012). If a new process was introduced to convert the excessive cloud water to rainwater, the bias would be mitigated because rainwater can deposit to the ground quickly. A candidate of the new process is the condensational conversion of cloud drops to raindrops via CCIs (see Fig. 1), which is tested herein via a simple case. The present bin model, in contrast to Berry and Reinhardt (1974), represents
the condensational conversion of cloud drops to raindrops via CCIs. As a result, it replicates the fast rain initiation observed (i.e., rain can initiate within half an hour or less; see Fig. 9). The difference in rain initiation between the present bin model and Berry and Reinhardt (1974) suggests that, if a weather/climate model properly represents the condensational conversion of cloud drops to raindrops via CCIs, its bias of “too dense clouds” will be mitigated effectively.

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Data availability statement. The datasets of modeled rain and CCI initiation time are available at https://osf.io/4tsn3/.

APPENDIX

Implicit Integration of the Condensational Growth Equation

To simulate CCN activation accurately, the bin model solves the condensation growth equation, (10), with an implicit scheme. Following Zeng (2018), (10) is rewritten in terms of r as

$$\frac{dr}{dt} = \frac{2}{\rho_l (A_w + B_w)} \left( S_w - 1 - \frac{C_1}{r} + \frac{C_m r}{r^n + 1} \right)$$

(A1)

where $\rho_l$ is the density of liquid water. The equation is then discretized to

$$\frac{(r^{n+1})^2 - (r^n)^2}{\Delta t} = \frac{2}{\rho_l (A_w + B_w)} \left[ S_w - 1 - \frac{C_1}{r^{n+1}} + \frac{C_m}{(r^n + 1)^2} \right].$$

(A2)

where $r^n$ and $r^{n+1}$ represent r on time level n and n + 1, respectively. Given $r^n$, (A2) is solved for $r^{n+1}$ with the Newton’s iterative method.

When r is small, the surface and solute terms in (A2) are large and thus their r is represented on time level n + 1. Even though so, if the time step $\Delta t$ is too large, (A2) may have two solutions: one for haze droplet and the other for cloud drop. To avoid an artificial jump from a haze droplet to a cloud drop, a small value of $\Delta t$ (e.g., 0.05 s) is used so that CCN activation is simulated accurately.

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