Application of Morrison Cloud Microphysics Scheme in GRAPES_Meso Model and the Sensitivity Study on CCN’s Impacts on Cloud Radiation

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Abstract: In the present study, the Morrison double-moment cloud microphysics scheme including mass and droplet number concentration of water and ice clouds is implemented into the Chinese mesoscale version of the Global/Regional Assimilation and Prediction System (GRAPES_Meso). Sensitivity experiments of different cloud condensation nuclei (CCN) values are conducted to study the impacts of CCN on cloud microphysical processes and radiation processes in East China. The model evaluation shows that the simulated cloud liquid water path (CLWP) is consistent with that of the National Center for Environment Prediction (NCEP) reanalysis, and the cloud optical depth (COD) and effective radius of cloud water (Rc) are in agreement with those of the Moderate Resolution Imaging Spectroradiometer (MODIS) datasets both in regional distribution and magnitudes. These comparisons illustrate the effectiveness of the Morrison scheme for the cloud processes in East China. For the study period of 8 to 12 October 2017, the sensitivity experiments show that with initial CCN number concentration (CCN0) increasing from 10 to 3000 cm−3, the maximum value of daily average Rc decreases by about 63%, which leads to a decrease of cloud-rain conversion rate. Moreover, the maximum value of daily average mixing ratio of cloud water (q_c) increases by 133%, the maximum value of daily average mixing ratio of rain (q_r) decreases by 44%, and the maximum value of daily average CLWP and COD increase by 100% and 150%, respectively. This results in about 65% increasing of the maximum value of daily average cloud downward shortwave radiative forcing (CDSRF) when CCN0 increases from 10 cm−3 to 3000 cm−3. The study indicates the important impacts of CCN on cloud properties and radiation effects.

Keywords: numerical simulation; cloud microphysical processes; Morrison scheme; cloud condensation nuclei; cloud radiation

1. Introduction

Clouds are an essential component for the earth’s water cycle, and intensely influence energy exchange between earth and atmosphere [1,2]. The interaction between clouds and aerosols directly affects cloud properties and microphysical processes by radiation and heat
transfer, which then has important influences on weather and climate change. A reasonable cloud microphysics scheme describing the cloud micro and macro properties, formation, and evolution accurately are vital for the weather and climate model [3,4].

Cloud microphysics parameterization schemes are generally divided into bin schemes [5,6] and bulk schemes [7,8]. Bin schemes explicitly calculate the evolution of the particle size distribution and divide a hydrometeor size distribution into several size bins, based on the microphysical properties of hydrometeors and aerosol such as phase state, particle size, shape, density, which is widely used to study the liquid and ice-phase cloud microphysical processes [9,10]. Bulk schemes, therefore, become a feasible alternative for many applications. Bulk schemes can be further classified into single-moment and double-moment schemes. Single-moment schemes simulate only the mass mixing ratio of hydrometeors [11–13]. Double-moment schemes simulate both mass mixing ratio and number concentration of hydrometeors [14]. Double-moment microphysics schemes were widely used in the convective and layer clouds and precipitation [15,16]. Ferrier proposed a double-moment ice-phase volumetric water microphysics scheme to simulate the distribution of condensate in convective precipitation and layered cloud precipitation under different large-scale environmental conditions [17]. A relatively complete double-moment microphysics scheme was proposed to study cumulonimbus related issues, which can numerically calculate the number concentration of cloud ($n_c$), raindrops ($n_r$), ice ($n_i$), snow ($n_s$), and graupel ($n_g$) droplets [18]. Double-moment schemes incorporate the number concentration of hydrometeors and increase the degrees of freedom of the particle size distribution, and the number and quality of hydrometeor particles in the physical mechanism are more coordinated, which is incorporated in climate or weather models and widely used in weather and climate simulations [19–21].

Morrison developed a double-moment microphysics scheme including five types of hydrometeors: cloud droplets, raindrops, ice, snow, and graupel [22]. The scheme considers a more elaborate and comprehensive microphysical process, mass concentration, and numerical concentration of the aquatic matter and is used to test real cases and ideal case studies [23–26]. The scheme is widely applied to investigate the influence of aerosol spectrum changes on the precipitation weather process. Therefore, this double-moment scheme has been widely used in the popular mesoscale models such as the NCAR/Penn fifth-generation Mesoscale Model (MM5) and the Weather Research and Forecasting (WRF) model. The Morrison scheme is implemented into the aerosol-climate model BCC_AGCM2.0.1_CUACE/Aero to evaluate model results for aerosols cloud properties and meteorological fields, the new model simulated a more realistic aerosol mass concentrations and optical depth compared with the original version [21]. The forecast of cold precipitation amount includes cloud ice mixing ratio, snow mixing ratio, and haze mixing ratio; the Morrison scheme can also forecast the number concentration [27]. Besides, the predicted particle properties (P3) scheme has been developed, and the scheme represents ice microphysics by emphasizing the pre-diction of particle properties rather than the separation of ice into different predefined categories [28]. Thompson had also developed a new bulk microphysical parameterization (BMP) to analyze the impact of cloud properties on radiation, precipitation amount and type, and aircraft icing, which has been updated to incorporate aerosols explicitly in a simple and cost-effective manner [29,30]. A comparative study of different microphysical schemes (Lin, WSM6, GCE, Thompson and Morrison) was carried out, and it was found that although the Thompson scheme and Morrison scheme set the cloud droplet concentration as a constant, the cloud droplet number concentration and cloud droplet spectrum of these two schemes are closer to the observation [31–33].

Cloud condensation nuclei (CCN) plays a crucial role in the double moment cloud microphysical processes. The CCN may influence the precipitation efficiency by determining the concentration and original particle size of the cloud droplet. Studies show that the increase of CCN will lead to an increase in the concentration of cloud droplets and a decrease in the size of cloud droplet particles, which then suppresses warm cloud precipitation [34,35]. Besides, with the increase of CCN, the mixing ratio of cloud water
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(qc) in the condensate increases, the size of droplets decreases, and raindrops decrease, accordingly [36]. CCN can also affect the cloud radiative forcing through the indirect effect [37]. That is, the increase of CCN will lead to more and smaller cloud droplets, which then generates more cloud reflection. The effective radius of cloud water (Rc) is an important cloud physical characteristic parameter, and it can affect the scattering characteristics of clouds. For a given liquid water content or ice water content, cloud with smaller Rc or the effective radius of cloud ice (Ri) will reflect/scatter more solar radiation [38]. The distribution of the cloud optical depth (COD) is affected by cloud volume, cloud water content, and Rc. The area with large cloud volume is basically the large value area of COD. From the perspective of vertical change, the change of COD is greatly affected by the change of liquid water content with height [39].

These studies on CCN are mainly concentrated in areas where aerosol pollution is relatively light, while for the heavily polluted North China Plain, research about the impact of CCN on cloud microphysical processes is still poor. In this study, the Morrison double-moment cloud microphysics scheme is implemented into the GRAEPS_Meso model. The evaluation of the simulation results and the impacts of different CCN on microphysical quantities and cloud radiation forcing have been presented. Table 1 shows the related acronyms of cloud microphysical characteristics used in this paper, such as CCN, Rc, Ri, and so on. The study indicates the impacts of CCN on cloud properties and radiation effects are important. There is high pollution in East China, and the number concentration of CCN changes drastically. The results of my study are very important for the improvement of forecasting ability and the model performance in East China.

Table 1. List of the acronyms used.

| Notation | Description |
|----------|-------------|
| CCN      | cloud condensation nuclei |
| Rc       | effective radius of cloud water |
| Ri       | effective radius of cloud ice |
| qc       | mixing ratio of cloud water |
| qr       | mixing ratio of cloud raindrops |
| qS       | mixing ratio of cloud snow |
| qι       | mixing ratio of cloud ice |
| qιS      | mixing ratio of cloud graupel |
| nc       | number concentration of cloud water |
| nr       | number concentration of raindrops |
| ns       | number concentration of snow |
| ni       | number concentration of ice |
| ng       | number concentration of graupel |
| CLWP     | cloud liquid water path |
| COD      | cloud optical depth |
| CWPC     | cloud liquid water path of cloud water |
| CWPI     | cloud liquid water path of cloud ice |
| CODC     | cloud optical depth of cloud water |
| CODI     | cloud optical depth of cloud ice |
| CDSRF    | cloud downward shortwave radiative forcing |

2. Model and Scheme

2.1. GRAPESMeso

The mesoscale version of the new-generation Global/Regional Assimilation and Prediction System (GRAPESMeso) is an operational weather forecasting model developed by the Chinese Academy of Meteorological Sciences, which mainly consists of 3-D meteorological field data assimilation, the full compressible dynamic framework, and a physical parameterization package [40–42]. This model is generally used for operational forecasting and scientific research about the medium- and short-term weather conditions in China [43,44]. It has also been widely used for the study of cloud, aerosol, and the interaction between cloud and aerosol in China for about 20 years [45,46].
results of the GRAPES_Meso model in national and regional areas are satisfactory [47,48]. The model adopts a height-based terrain-following coordinate, a semi-implicit and semi-Lagrangian temporal advection scheme for time integration discretization, an Arakawa-C staggered grid format, a central finite-difference scheme for horizontal discretization, and a non-hydrostatic approximation scheme for vertical discretization [49]. The physical parameterization schemes selected in this study include Rapid Radiative Transfer Model (RRTM) longwave radiation scheme [50], Goddard shortwave radiation scheme [51], the Monin–Obukhov surface layer scheme [52], the Noah land surface model [53], the Betts–Miller–Janjic (BMJ) cumulus convection scheme [54], and the Medium-Range Forecast (MRF) boundary layer scheme [55].

2.2. Morrison Cloud Microphysical Scheme

Morrison double-moment microphysics scheme including mass mixing ration of and number concentration predictions of hydrometeors particles is integrated into GRAPES_Meso to simulate the cloud process in East Asia in this study [56]. The governing equations of the Morrison scheme for the mass mixing ratio and number concentration of hydrometeors are as follows [57,58]:

\[
\frac{\partial \theta}{\partial t} = -\nabla \cdot (\theta \mathbf{v}) + \frac{\partial}{\partial x} (V_{\theta x} \theta) + \nabla D \theta + \left( \frac{\partial \theta}{\partial x} \right)_{\text{COND/DEP}} + \left( \frac{\partial \theta}{\partial x} \right)_{\text{AUTO}} + \left( \frac{\partial \theta}{\partial x} \right)_{\text{COAG}} + \left( \frac{\partial \theta}{\partial x} \right)_{\text{MLT/FRZ}} + \left( \frac{\partial \theta}{\partial x} \right)_{\text{MULT}}
\]

\[
\frac{\partial N}{\partial t} = -\nabla \cdot (N \mathbf{v}) + \frac{\partial}{\partial x} (V_{N x} N) + \nabla D N + \left( \frac{\partial N}{\partial x} \right)_{\text{PRO}} + \left( \frac{\partial N}{\partial x} \right)_{\text{EVAP/SUB}} + \left( \frac{\partial N}{\partial x} \right)_{\text{AUTO}} + \left( \frac{\partial N}{\partial x} \right)_{\text{SELF}} + \left( \frac{\partial N}{\partial x} \right)_{\text{COAG}} + \left( \frac{\partial N}{\partial x} \right)_{\text{MLT/FRZ}} + \left( \frac{\partial N}{\partial x} \right)_{\text{MULT}}
\]

where \( q \) and \( N \) represent the mass mixing ratio and number concentration of all hydrometeors; they depend on space variables \( x, y, \) and \( z \) and time, \( t \). \( \mathbf{v} \) is the three-dimensional wind vector. \( V_{\theta x} \) and \( V_{N x} \) are number- and mass-weighted the final falling velocity of each particle. \( \nabla D \) is a turbulent diffusion operator for models that parameterize turbulent mixing. The first three terms on the right-hand side of Equations (1) and (2) represent the advection, sedimentation, and the turbulent diffusion term of \( q \) and \( N \), respectively, and they are calculated by dynamic frame in the GRAPES_Meso model. Table 2 summarized all the transformation processes in Equations (1) and (2) [22].

| Process | Description |
|---------|-------------|
| PRO     | Ice nucleation or droplet activation from aerosol |
| COND/DEP| Condensation of droplets and rain/deposition of snow and cloud ice (evaporation of droplets and rain/sublimation of snow and cloud ice) |
| AUTO    | Autoconversion (parameterized transfer of mass and number concentration from the cloud ice and droplet classes to snow and rain due to coalescence and diffusional growth) |
| COAG    | Collection between hydrometeor species (droplets, cloud ice, snow, and rain) |
| MLT/FRE | Melting of snow to form rain and cloud ice to form droplets/freezing of droplets and rain to form cloud ice and snow |
| EVAP/SUB| Evaporation of rain and droplets/sublimation of cloud ice and snow |
| SELF    | Self-collection of droplets, cloud ice, snow, and rain |
| MULT    | Ice multiplication (transfer of mass from the snow class to ice) |

The Morrison scheme includes not only the double-moment of warm cloud processes but also the double-moment of cold cloud processes. It can predict water cloud mass mixing ration, number concentration \( (q_c, n_c) \), mixing ratio of rainwater \( (q_r, n_r) \), cloud mass mixing ratio, and number concentration (mixing ratio of ice \( (q_i, n_i) \), \( q_g \), mixing ratio of snow \( (q_s, n_s) \), \( n_g \)). The scheme includes 19 microphysics processes mass mixing ratio related to mass mixing ratio and 22 microphysics processes related to number concentration, respectively. The scheme uses the activation calculation method to calculate CCN, while this study uses logarithmic aerosol particle size distribution to drive the CCN spectrum. Therefore, it needs to preset an initial CCN number concentration \( (CCN_0) \) as the assumed atmosphere.
condition [59]. The default setting of CCN$_0$ is 250 cm$^{-3}$, representing the typical continent cloud, and many studies use this fixed default values in the simulation due to the lack of aerosols activation scheme in most numerical weather prediction models and cloud simulation studies, which is one of the major reasons leading to model uncertainties from clouds features and processes.

2.3. Experiments Design

October is fall in China, and the frontal cloud system featured by stable stratus is the typical cloud system in this region. There are two frontal cloud systems passed through the study region in October. One occurred 8–12, another occurred 23–28; they are both frontal cloud systems dominated by stable stratus instead of convective clouds moved from northwest to southeast in East China, and the convective cloud is extremely weak during the cloud processes. The observation studies showed that the CCN varies largely in different regions and atmosphere conditions closely related to atmosphere critical supersaturation, aerosol loading, and species. It was reported that CCN could reach up to 1000 and 40,000 cm$^{-3}$ in different conditions [60,61]. In North China, when the heaviest aerosol pollution occurs, the maximum value of CCN can get to 8000 cm$^{-3}$, and when the pollution is light, the minimum value of CCN can get to 8.2 cm$^{-3}$ [62,63]. However, the initial values are usually set as fixed 250 cm$^{-3}$ in the microphysics scheme in weather and climate models, which will lead to uncertainties in cloud radiation processes and then the model performance [36,64]. This study incorporates the Morrison scheme with the GRAPES_Meso model and sets the initial CCN as 250 cm$^{-3}$ to evaluate its applicability in the cloud system simulation in east Asia. The cloud liquid water path (CLWP) of reanalysis from the National Center for Environment Prediction (NCEP), COD and Rc of level-2 products from the Moderate Resolution Imaging Spectrometer (MODIS) are used for the model evaluation. Based on this, a set of sensitivity experiments CCN$_0$ are set as 10, 100, 250, 600, 1000, 3000, 8000, and 10,000 cm$^{-3}$ to study the effects of CCN on the cloud properties and radiation processes. The increase of CCN will lead to an increase in cloud droplets, which has been fully proved, so CCN and cloud droplets are in common to a certain degree, especially in variations. In this paper, we apply the Morrison double-moment scheme in the GRAPES model. However, in the current model, the processes of CCN activation have not been realized. We just set a wide range of possible CCN values directly (named as CCN$_0$) to study the impacts of possible CCN’s changing on clouds radiation. Similar experiments have been conducted in previous studies [65,66]. Our following paper will focus on the different CCN activation schemes and its impacts on clouds radiation. For the model configuration, the initial input and lateral boundary are provided by NCEP final analysis (FNL) data (https://rda.ucar.edu/datasets/ (accessed on 20 July 2019)), with 0.25$^\circ$ × 0.25$^\circ$ horizontal grids and 6 h interval. The model domain covers east China, and its downwind area (20$^\circ$–50$^\circ$ N, 100$^\circ$–135$^\circ$ E). The model has 31 vertical layers from the surface to about 30 km. The model starting time is 00 UTC 8 October 2017, and its ending time is 00 UTC 13 October 2017, and the study period is from 00 UTC 8 to 12 October.

3. Model Evaluation

The CCN$_0$ in Morrison scheme is assumed as the background of continent atmospheric condition 250 cm$^{-3}$ in the model evaluation; in contrast, CCN of 100 and 300 cm$^{-3}$ are also used for comparison in this section. Compared with WDM6 scheme in GRAPES_Meso, Morrison scheme has better performance in simulating the cloud microphysical characteristics in this study (Figures S1–S3). The correlation coefficient (CORR) and Root Mean Squared Error (RMSE) of CLWP, Rc, and COD between observations and simulations of Morrison and WDM6 schemes with setting CCN values as 250 cm$^{-3}$ are calculated in the supplementary material (Table S1); it also can be seen that the simulations of Morrison scheme are better than those of WDM6 scheme.

Figure 1a shows the modeled average Rc, COD, and CLWP (CCN set as 100, 250, and 300 cm$^{-3}$) from 8 to 12 October 2017 and cloud MODIS Rc, COD, and CLWP of
NCEP Analysis. The comparison of modeled Rc with that of MODIS shows that there are obvious differences among the modeled Rc when the CCN is set as 100, 250, and 300 cm\(^{-3}\), indicating the great impacts of CCN on clouds particle size. In general, modeled Rc is the most consistent with the MODIS Rc when CCN is 250 cm\(^{-3}\) while Rc is generally bigger when CCN is 100 cm\(^{-3}\) and smaller when CCN is 300 cm\(^{-3}\) than those of MODIS. This is because more CCN means more cloud particles sharing the same cloud water and the reduction of the particles’ size. The size reduction of cloud particles with the CCN increasing causes the decreasing of the auto transformation process from clouds to rain, which further results in more cloud water in the atmosphere, so modeled COD and CLWP are both increased with the CCN decreasing from 100, 250 to 300 cm\(^{-3}\), suggesting the possible impacts of CCN on cloud radiation by Rc, COD, and CLWP. Moreover, the two parameters agree the best with those between MODIS COD and NCEP CLWP when CCN is 250 cm\(^{-3}\), illustrating the basic rationality of 250 cm\(^{-3}\) when CCN has to be set as a constant when aerosols activation scheme and aerosols processes are absent in the numerical weather prediction (NWP) models. We also compare the results between model and observed data by using the case from 23 to 28 October 2017 (Figures S4–S6). When the initial CCN is set as 250 cm\(^{-3}\), simulations of Rc, COD and CLWP agree with those of MODIS and NCEP in space and time. CORR and RMSE of CLWP, Rc, and COD between observations and simulations with setting CCN values as 250 cm\(^{-3}\) are calculated in Table 3. In the case from 8 to 12 October 2017, the CORRs of the observed and simulated RC, COD, and CLWP are 0.54, 0.55, and 0.57, and the RMSEs are 8.9, 1.6, and 160.4. As for another case from 23 to 28 October 2017, the CORRs are 0.51, 0.48, and 0.55, and the RMSEs are 10.2, −2.1, and 173.8. All of CORRs have exceeded 0.05 significance level. These results show that the simulations when the initial CCN is set as 250 cm\(^{-3}\) agree with the observed data and are more accurate than those when setting CCN as 100 cm\(^{-3}\) or 300 cm\(^{-3}\). However, it is noteworthy that there are certain diversities between modeled Rc, COD, and CLWP even CCN is 250 cm\(^{-3}\) and those of MODIS and NCEP. The truth of the obvious differences between Rc, COD, and CLWP among the three experiments when CCN is 100, 250 or 300 cm\(^{-3}\) indicates the important impacts of CCN on cloud key parameters. The CCN is set as the constant of 250 cm\(^{-3}\) in Morrison in the whole domain during model running time maybe still one important reason leading to the model errors of Rc, COD, CLWP, and cloud process though it is the best choice at present comparing with the model results when CCN is 100 and 300 cm\(^{-3}\).

Table 3. Comparison of observed and simulated average Rc, COD and CLWP with CCN values of 250 cm\(^{-3}\).

|                      | from 8 to 12 October 2017 | from 23 to 28 October 2017 |
|----------------------|---------------------------|---------------------------|
|                      | CORR | RMSE | CORR | RMSE |
| Rc (µm)              | 0.54 | 8.9  | 0.51 | 10.2 |
| COD                  | 0.55 | 1.6  | 0.48 | −2.1 |
| CLWP (g/m\(^2\))    | 0.57 | 160.4| 0.55 | 173.8|
4. Study Results and Discussions

To illustrate the possible impacts of CCN\textsubscript{0} on clouds physical, microphysics, and optical features, the radiation processes, temporal and spatial changing of mass mixing of cloud hydrometeors \((q_c, q_r, q_i, q_s, q_g)\), \(R_c\), \(R_i\), COD of water cloud \((\text{CODC})\), and ice cloud \((\text{CODI})\) corresponding to different CCN\textsubscript{0} values in Morrison scheme, and the possible relationships between the changes of above factors are compared and discussed in detail in this section.

4.1. The CCN Impacts on the Mass Mixing Ratio of Hydrometeors

Firstly, Figure 2 gives the vertical distribution of the average mixing ratio five kinds of hydrometeors, \(q_c, q_r, q_i, q_s,\) and \(q_g\) simulated by the Morrison scheme with CCN\textsubscript{0} values of 10 cm\textsuperscript{-3}, representing CCN in a clean atmosphere, 250 cm\textsuperscript{-3} representing typical continent CCN, and 3000 cm\textsuperscript{-3} representing CCN in the severe polluting atmosphere from 8 to 12 October 2017. The black line is the 0\(^\circ\) temperature line at the height of about 600 hPa.
It can be seen from Figure 2 that $q_c$ gets to the maximum value from 04:00 UTC to 19:00 UTC on 9 October from the height of 800 hPa to 650 hPa with CCN$_0$ values of 10 cm$^{-3}$. The $q_c$ gets to the maximum value at a height of 600 hPa from 6:00 UTC to 18:00 UTC on 9 October with CCN$_0$ values of 250 cm$^{-3}$. With values of CCN$_0$ varying to 3000 cm$^{-3}$, $q_c$ increases...
significantly, and the increase in CCN₀ enhances q_c by 133%. The q_r gets to the maximum value after 11:00 UTC on 9 October at the height of 950 hPa to 750 hPa; q_r decreases with values of CCN₀ increasing to 250 cm⁻³, and the ground gets to a maximum value at the height from land layer to 700 hPa during the period of 13:00 UTC on 9 October to 02:00 UTC on 10 October. With the values of CCN₀ increasing to 3000 cm⁻³, q_r has a large reduction significantly. After 14:00 UTC on 9 October, q_r decreases by 44% compared with 10 cm⁻³. Figure 3 displays the vertical profiles of the region and period averaged q_c, q_r, q_i, q_s with CCN₀ of 250 cm⁻³ (a) and the differences between it and that of CCN₀ of 10 cm⁻³ (b) and 3000 cm⁻³ (c). It can be seen clearly that comparing Figure 3a–c when CCN₀ is set as 10 cm⁻³, q_c decreases and q_r increases comparing with those when CCN₀ is 250 cm⁻³; the maximum of the q_c differences between the two is up to 10 mg/kg, which is an account for 43% of the maximum value of q_c when CCN₀ is 250 cm⁻³. Correspondingly, the maximum of q_r decrease is 6, accounting for almost 100% of q_r when CCN₀ is set as 250 cm⁻³. Conversely, when CCN₀ is set as 3000 cm⁻³, q_c increases and q_r decreases comparing with those when CCN₀ is 250 cm⁻³. The maximum of the q_c differences between the two is about 5 mg/kg, which means about a 22% increase of the maximum value of q_c when CCN₀ is 250 cm⁻³, and the maximum q_r decrease is about 20%.

![Figure 3. Vertical profiles of area-averaged q_c, q_r, q_i, q_s and q_g (mg/kg) of A region from 8 to 12 October (a) CCN₀ = 250 cm⁻³, (b) the difference between CCN₀ = 10 cm⁻³ and CCN₀ = 250 cm⁻³, (c) the difference between CCN₀ = 3000cm⁻³ and CCN₀ = 250 cm⁻³).](image-url)

However, comparing with q_c and q_r, the changes of q_i and q_s due to the CCN₀ changing is small (Figure 2). The sources of cloud ice crystals are mainly from the ice nucleation from freezing of aerosol and the heterogeneous freezing of droplets, among which ice nucleation from the freezing of aerosol is more important [16,22]. When CCN number concentration increases, the impact on the ice nuclei is little, and the largest source of ice crystal is not changed [67]. Moreover, because the cloud system studied here is a stratus cloud with few ascending motions, it is difficult to freeze a large amount of cloud water. Based on these, the number concentration of CCN changes will not change the ice crystals much. Meantime, the source of snow and graupel is ice crystals [68]. Therefore, compared with cloud water and cloud raindrops, the changes of cloud ice, cloud snow, and graupel are not obvious. Although this change is small, the mixing ratio of cloud snow and graupel also changes to a certain extent with the increase of the CCN concentration, which is manifested as a rise near and below the 0 °C isotherm and a drop in the upper air slightly (Figure 3). This result agrees with the studies that the increase of CCN is not conducive to the formation of ice particles in the upper air [16–19,22,36,37]. The reason may be that the increase of cloud droplets consumes ice crystals and causes them to fall above 0 °C isotherm. In addition, the increase of CCN causes the cloud water to rise significantly near the 0 °C isotherm, which is beneficial to the growth of the snow and graupel. Therefore, the mixing ratio of cloud snow and graupel increase with CCN increasing below 500 hPa.
4.2. CCN Impacts on Modeled $R_c$, CLWP, and COD

CLWP represents the total cloud water atmosphere column loading and has an important influence on the total cloud radiation process. $R_c$, the cloud particle size, is the very sensitive parameter on the cloud optical features and on the cloud radiation process. COD is the direct input parameter combining the contributions of cloud water loading and optical impacts in the radiative transfer model to calculate the cloud radiation process [64].

Figure 4 shows the simulated daily $R_c$ from 8 October to 12 October when $C_{CN0}$ is set as 10, 250, and 3000 cm$^{-3}$ in Morrison scheme. It can be seen from Figure 4 that there are distinguished differences among the simulated $R_c$ of the three $C_{CN0}$ values. In general, $R_c$ decreases with the $C_{CN0}$ increasing, and $R_c$ is the biggest when $C_{CN0}$ is set as 10 cm$^{-3}$; the second corresponds to $C_{CN0}$ of 250 cm$^{-3}$, and the smallest corresponds to $C_{CN0}$ of 3000 cm$^{-3}$. Although the $R_c$’s differences between $C_{CN0}$ of 10 cm$^{-3}$ and 250 cm$^{-3}$, representing the possible $R_c$’s differences between very clean and normal continent $C_{CN0}$, are much bigger than those between $C_{CN0}$ of 250 cm$^{-3}$ and 3000 cm$^{-3}$, representing the $R_c$’s differences between normal continent $C_{CN0}$ and $C_{CN0}$ of severe pollution atmosphere, the $R_c$’s differences between $C_{CN0}$ of 250 cm$^{-3}$ and 3000 cm$^{-3}$ are still obvious enough to show the possible reforming on $R_c$ due to the $C_{CN0}$ changing from normal continent atmosphere to severe pollution atmosphere. The maximum value of daily average $R_c$ decreases by about 63% when $C_{CN0}$ increases from 10 cm$^{-3}$ to 3000 cm$^{-3}$. It is worth noting that for CLWP (Figure 5), the changes in CLWP are consistent with $C_{CN0}$, but the magnitude of increase in CLWP is much smaller than the increase in $C_{CN0}$ with getting smaller tendency. In this case, in which the increase in cloud water content is relatively low, too many cloud droplets compete for water and are not prone to growth, which will lead to rapid decrease in $R_c$ and the magnitude of the decrease decreases [69].

Figure 5 displays the daily average CLWP by the model from 8 to 12 October when $C_{CN0}$ is 10, 250, and 3000 cm$^{-3}$ in Morrison scheme in the model run. It can be seen that CLWP corresponding $C_{CN0}$ of 10 cm$^{-3}$ is the smallest, CLWP corresponding $C_{CN0}$ of 250 cm$^{-3}$ is the second, and CLWP corresponding $C_{CN0}$ of 3000 cm$^{-3}$ is the biggest, illustrating that more $C_{CN0}$ results in less $R_c$ (Figure 4) and the automatic conversion process from cloud to rain, and more cloud water exits in the atmosphere. The maximum value of daily average CLWP increases by 100% when the values of $C_{CN0}$ going from 10 cm$^{-3}$ to 3000 cm$^{-3}$. Comparing the CLWPs under the three $C_{CN0}$ values, it can be found that the CLWP differences between $C_{CN0}$ as 10 and 250 cm$^{-3}$ are bigger than those between $C_{CN0}$ as 250 and 3000 cm$^{-3}$ under the same background of atmospheric circulation and water vapor. Model daily COD (Figure 6) shows a similar change with the $C_{CN0}$ changing, that is, COD with $C_{CN0}$ as 10 cm$^{-3}$ is the smallest, COD with $C_{CN0}$ as 250 cm$^{-3}$ is the middle, and COD with $C_{CN0}$ as 3000 cm$^{-3}$ is the biggest. The maximum value of daily average COD increases by 150% when the values of $C_{CN0}$ go from 10 cm$^{-3}$ to 3000 cm$^{-3}$. COD has a good positive correlation with CLWP. The bigger the cloud water content is, the greater the cloud optical depth is, which causes the less solar shortwave radiation received by the ground [69]. However, there is still a certain difference between the CLWP and COD changing with the $C_{CN0}$, that is, COD changes more with $C_{CN0}$ from 250 to 3000 cm$^{-3}$ than CLWP did. This suggests that COD increases much more with $C_{CN0}$ changing from normal continent (250 cm$^{-3}$) to severe pollution $C_{CN0}$ (3000 cm$^{-3}$) than CLWP does. This can be interpreted as that CLWP is only changed by the changes of cloud water mixing by $C_{CN0}$, but COD may be changed by both the changes of cloud water mixing and optical properties of cloud particles due to the cloud size changing. When $C_{CN0}$ changes from 10 to 250 cm$^{-3}$, the cloud particle size is relatively big; the impact of $C_{CN0}$ on cloud water content is bigger than that on optical features of cloud particles, and thus the CLWP difference is bigger than COD. When $C_{CN0}$ changes from 250 to 3000 cm$^{-3}$, the cloud particle size is much smaller; the impact of $C_{CN0}$ on optical features of cloud particles is much more important than it on cloud water content, and thus, the COD difference is bigger than CLWP. This result suggests the possible COD difference due to normal continent $C_{CN0}$ and severe pollution $C_{CN0}$. 
Figure 4. Daily averaged Rc (µm) by model from 8 to 12 October (left: CCN$_0$ = 10 cm$^{-3}$, mid: CCN$_0$ = 250 cm$^{-3}$, right: CCN$_0$ = 3000 cm$^{-3}$).
Figure 5. Daily averaged CLWP (g m$^{-2}$) by model from 8 to 12 October (left: CCN$_0$ = 10 cm$^{-3}$; mid: CCN$_0$ = 250 cm$^{-3}$; right: CCN$_0$ = 3000 cm$^{-3}$).
The above discussion uses three CCN$_0$ values of 10 (representing very clean atmosphere), 250 (representing continent atmosphere), 3000 cm$^{-3}$ (representing severe pollution atmosphere) in the Morrison scheme to discuss the CCN impacts on cloud properties. However, in the real atmosphere, CCN varies with meteorological conditions and aerosols types and concentrations. The experiment results when CCN is set as 10, 100, 250, 600, 1000, 3000, or 8000 cm$^{-3}$ will be used to discuss the different CCN values on cloud properties in detail. Hence, Region A (32°–42° N, 105°–130° E) is chosen to study the effect of different values of CCN on area-averaged cloud microphysical quantities.

Figure 7 displays the temporal changing of region averaged $R_c$, $R_i$, CODC, CODI,
CLWP of cloud water (CWPC), and CLWP of cloud ice (CWPI) corresponding to the different CCN$_0$ from 8 to 12 October. It can be seen that Rc (Figure 7a) decreases, while CODC (Figure 7c) and CWPC (Figure 7e) increase with the CCN$_0$ increasing from 10, 100, 250, 600, 1000, 3000, to 8000 cm$^{-3}$. This is mainly because more CCN in the atmosphere leads to a smaller size of cloud particles (smaller Rc), less auto conversion from cloud to rain, more water cloud with smaller particle sizes in the atmosphere, and higher atmospheric column cloud water, which means bigger CODC and CWPC. Among the Rc differences between every two adjacent CCN$_0$ values, the difference between Rc when CCN$_0$ is set as 10 cm$^{-3}$ and that when CCN$_0$ is 250 cm$^{-3}$ is the biggest. Though CODC and CWPC show similar changes with the CCN$_0$, there are still some differences: Similar to Rc changes, the difference between CWPC when CCN$_0$ is set as 10 cm$^{-3}$ and that when CCN$_0$ is 250 cm$^{-3}$ is the biggest among the CWPC differences between every two adjacent CCN$_0$ values. CWPC changes with CCN$_0$ from 100, 250, 600, 1000, 3000, to 8000 cm$^{-3}$ is not as large as it is with CCN$_0$ from 10 to 100 cm$^{-3}$. However, CODC changing with CCN$_0$ shows certain different features to those of Rc and CWPC, that is, the largest differences in CODC between per two adjacent CCN$_0$ occur in CCN$_0$ changing from 1000 to 3000 cm$^{-3}$ or 3000 to 8000 cm$^{-3}$, instead of CCN$_0$ changing from 10 to 100 cm$^{-3}$ as Rc and CWPC do. In other words, when atmosphere CCN changes from a very clean atmosphere (10 cm$^{-3}$) to continent atmosphere (100 or 250 cm$^{-3}$), Rc and CWPC change the most, and COD changes most with CCN continued increases based on pollution atmosphere (from 1000 to 3000, or 8000 cm$^{-3}$). Furthermore, COD changes uniformly with CCN$_0$ changing from 10, 100, 250, 600, 1000, 3000 to 8000 cm$^{-3}$ while CLWP and Rc change most with CCN$_0$ changing from 10 to 100 cm$^{-3}$. This is because that CWPC is only affected by atmosphere column cloud water, but COD is affected by both atmosphere column cloud water and optical features of cloud particle sizes. Comparing with water cloud, ice cloud parameters including Ri (Figure 7b), CODI (Figure 7e), and CWPI (Figure 7f) change slightly with CCN$_0$.

Figure 7. Cont.
4.3. CCN Impacts on Cloud Radiation Forcing

The discussion in the above section illustrates the important impacts of CCN changing on cloud features, especially on water cloud features, which is sure to lead the changes in the cloud radiation process. The increase in number concentration and the decrease in size of cloud droplets will lead to increase of cloud albedo (Twomey effect). In addition, the decrease in particle size will reduce the precipitation rate and increase the liquid water content in the cloud, the thickness of the cloud, and the life of the cloud (Albrecht effect). These processes will change cloud albedo, resulting in the decrease of solar shortwave radiation received by the ground [3,70]. The cloud downward shortwave radiative forcing (CDSRF) provides a means to quantify the effect of cloud on radiation. The CDSRF is calculated by the Equation (3):

\[
\text{CDSRF} = \downarrow F_{\text{sw, clear}}^{\text{down}} - \downarrow F_{\text{sw, cloudy}}^{\text{down}}
\]  

where \(\downarrow F_{\text{sw}}^{\text{down}}\) is the simulated downward shortwave radiation received by the surface. The subscripts “clear” and “cloudy” refer to clear-sky and cloudy (all-sky) conditions.

Figure 8 shows CDSRF at 12:00 UTC from 8 to 12 October 2017 when CCN\(_0\) is 250 \(\text{cm}^{-3}\) and the difference between it and that when CCN\(_0\) is 10 \(\text{cm}^{-3}\) or 3000 \(\text{cm}^{-3}\). It can be seen that there are distinguished differences among the simulated CDSRF of the three CCN\(_0\) values. In general, CDSRF with CCN\(_0\) as 10 \(\text{cm}^{-3}\) is the smallest, CDSRF with CCN\(_0\) as 250 \(\text{cm}^{-3}\) is the middle, and CDSRF with CCN\(_0\) as 3000 \(\text{cm}^{-3}\) is the biggest. When CCN\(_0\) is set as 10 \(\text{cm}^{-3}\), CDSRF increases comparing with that when CCN\(_0\) is 250 \(\text{cm}^{-3}\); the maximum of the CDSRF differences between the two is up to \(-220 \text{W/m}^2\), which decrease by about 37% compared with the maximum value of CDSRF when CCN\(_0\) is 250 \(\text{cm}^{-3}\). Conversely, when CCN\(_0\) is set as 3000 \(\text{cm}^{-3}\), the maximum of the CDSRF differences between the two is about 60 \(\text{W/m}^2\), which means about 10% increasing of the maximum value of CDSRF when CCN\(_0\) is 250 \(\text{cm}^{-3}\). Comparing the CDSRFs under the three CCN\(_0\) values, it can be found that the possible CDSRF differences between very clean and normal continent CCN\(_0\) are much bigger than CDSRF differences between normal continent CCN\(_0\) and CCN\(_0\) of severe pollution atmosphere. The maximum value of daily average CDSRF differences between CCN\(_0\) of 10 \(\text{cm}^{-3}\) and 3000 \(\text{cm}^{-3}\) is up to 260 \(\text{W/m}^2\), which means about a 65% increasing of the maximum value of daily average CDSRF when CCN\(_0\) is 10 \(\text{cm}^{-3}\).
Figure 8. Daily CDSRF (W/m²) at 12:00 UTC from 8 to 12 October 2017 (left: CCN₀ = 250 cm⁻³, mid: difference between CCN₀ = 10 cm⁻³ and CCN₀ = 250 cm⁻³, right: difference in values between CCN₀ = 3000 cm⁻³ and CCN₀ = 250 cm⁻³).

Figure 9 displays hourly area-averaged COD (Figure 9a) and CDSRF (Figure 9b) of Region A at 12:00 UTC corresponding to the different CCN₀ on 9 October 2017. It can be seen that COD and CDSRF increases with the CCN₀ increasing from 10, 100, 250, 600, 1000, 3000, to 8000 cm⁻³. The area-averaged COD and CDSRF get to the maximum value at 12:00 UTC on 9 October; this is mainly because higher COD leads to higher CDSRF. Among the CDSRF differences between every two adjacent CCN₀ values, the differences between CDSRF when CCN₀ is set as 10 cm⁻³ and that when CCN₀ is 100 cm⁻³ is the biggest. However, COD changing with CCN₀ shows a certain different feature with those of CDSRF, that is, the largest differences in COD between per two adjacent CCN₀ occur when CCN₀ changes from 3000 to 8000 cm⁻³, instead of those in CDSRF when CCN₀ changes from 10 to 100 cm⁻³. In other words, when atmosphere CCN changes from 10 to 100 or 250 cm⁻³,
CDSRF changes the most, while COD changes most when CCN continues to increase from 1000 to 3000 or 8000 cm\(^{-3}\).

Table 4 displays area-averaged CDSRF of Region A with CCN changing and their percentage changes compared with CCN\(_0\) (250 cm\(^{-3}\)) at 12:00 UTC from 8 to 12 October. CDSRF changes the most with CCN\(_0\) changing from 10 to 250 cm\(^{-3}\) on 9 October, and the maximum of the area-averaged CDSRF differences between the two is up to \(-80.8\) W/m\(^2\), which decrease by 28.9% compared with the maximum value of CDSRF when CCN\(_0\) is 250 cm\(^{-3}\). The maximum percentage change is up to 40.6% between 10 cm\(^{-3}\) and 250 cm\(^{-3}\) on 8 October, while the minimum of that is 3% between 250 cm\(^{-3}\) and 600 cm\(^{-3}\) on the same day. When atmosphere CCN changes from very clean atmosphere (10 cm\(^{-3}\) to continent atmosphere (250 cm\(^{-3}\)), the percentage change compared to CDSRF with CCN\(_0\) set as 250 cm\(^{-3}\) is between 23.0% and 40.6%; when the CCN\(_0\) changing from normal continent atmosphere (250 cm\(^{-3}\)) to severe pollution atmosphere (3000 cm\(^{-3}\)), the percentage change is between 6.5% and 11.6%. Particularly, the values of area-averaged CDSRF of Region A in Table 1 are smaller than those in Figure 8, this is because the distribution of daily average CDSRF is uneven in Region A.

Table 4. Area-averaged CDSRF (W/m\(^2\)) of Region A with CCN changing and their percentage change based on that with CCN\(_0\) set as 250 cm\(^{-3}\) at 12:00 UTC from 8 to 12 October.

| CCN\(_0\) (cm\(^{-3}\)) | October 8 | COD / Percentage Change | October 9 | October 10 | October 11 | October 12 |
|-------------------------|-----------|-------------------------|-----------|------------|------------|------------|
| 10                      | 133.4/-40.6% | 199.4/-28.9% | 134.8/-36.8% | 139/-26.2% | 94.1/-23.0% |
| 100                     | 179.2/-4.5%  | 266.5/-4.9%  | 198.8/-6.8%  | 179.3/-4.8% | 110.6/-9.4% |
| 250                     | 187.6       | 280.2       | 213.3       | 188.4       | 122.1       |
| 600                     | 193.3/3.0%   | 291.4/4.0%   | 221.1/3.7%   | 192.5/4.1%  | 129.2/5.8%  |
| 1000                    | 199.9/6.6%   | 293.6/4.8%   | 226.5/6.2%   | 196.6/4.4%  | 133.7/9.5%  |
| 3000                    | 202.8/8.1%   | 302.1/7.8%   | 235.2/10.3%  | 200.6/6.5%  | 136.3/11.6% |
| 8000                    | 205.3/9.4%   | 304.6/8.7%   | 239.8/12.4%  | 209.9/8.2%  | 138.6/13.5% |

5. Summary and Conclusions

The Morrison double-moment cloud microphysical scheme including mass and droplets number concentration of water and ice cloud is integrated into the GRAPES_Meso model and a large-area stratus cloud process from 00:00 UTC 8 October to 00:00 UTC 13 October 2017 is simulated by it. The MODIS Rc, COD, and NCEP reanalysis CLWP are used for model evaluation. Sensitivity experiments are performed with different values of CCN number concentration varying from 10, 100, 250, 600, 1000, 3000, to 8000 cm\(^{-3}\),
respectively, to study the effects of CCN on microphysics processes and cloud radiation effect in East China. The specific conclusions of the paper are summarized as follows.

Compared with WDM6 scheme in GRAPES_Meso, Morrison scheme has better performance in simulating the cloud microphysical characteristics in this study. The simulated CLWP shows a great agreement with that of NCEP reanalysis. Both the regional distribution and magnitude of simulated Rc and COD are consistent with those retrieved by MODIS satellite, indicating that the reasonability and availability of the Morrison scheme in the cloud description of cloud optical properties and radiation in GRAPES_Meso in East Asia.

With the values of CCN\(_0\) increasing from 10 cm\(^{-3}\) (representing a very clean atmosphere) to 3000 cm\(^{-3}\) (representing severe pollution atmosphere), the maximum value of daily average Rc decreases by 63%, while the impact of CCN has a little effect on Ri. Besides, the maximum value of the simulated daily average q\(_c\) increases by about 133%, the maximum value of daily average q\(_r\) decreases by 44%, the maximum value of daily average CLWP increases by 100%, and the maximum average COD increases by 150% while the CWPI and CODI decrease slightly. Because ice nuclei (the largest source of ice crystal) have not been changed, and the cloud system studied here is a stratus cloud with few ascending motions, it is difficult to freeze a large amount of cloud water. Based on these, the number concentration of CCN changes will not change the ice crystals much. Meanwhile, the source of snow and graupel are ice crystals. Therefore, compared with cloud water and cloud raindrops, the changes of cloud ice, cloud snow, and graupel are not obvious. When the number concentration of CCN\(_0\) exceeds 3000 cm\(^{-3}\), its impact on CLWP is small, but the impact on COD is big due to the combined impacts of CCN on cloud water and cloud optical features related to particle size. These processes will change cloud albedo when CCN\(_0\) increases, resulting in the decrease of solar shortwave radiation received by the ground. The maximum value of daily average CDSRF increases by 65% on 9 October. CDSRF changes the most when atmosphere CCN changes from 10 cm\(^{-3}\) to 100 or 250 cm\(^{-3}\). For the uneven distribution of daily average CDSRF in Region A, the values of area-averaged CDSRF of Region A are smaller than the values of daily averaged CDSRF. These study results show CCN’s important impacts on cloud properties and radiative forcing. Because of the complexity of the effect of CCN on clouds, further studies will focus on more cloud processes and types, more microphysical schemes, and CCN or ice cloud nucleation (IN) activation.

**Supplementary Materials:** The following are available online at https://www.mdpi.com/article/10.3390/atmos12040489/s1, Figure S1: Comparation between MODIS and CCN \(= 250\) cm\(^{-3}\) Rc from 23 to 28 October 2017, Figure S2: Comparation between MODIS and CCN \(= 250\) cm\(^{-3}\) COD from 23 to 28 October 2017, Figure S3: Comparation between NCEP and CCN \(= 250\) cm\(^{-3}\) CLWP from 23 to 28 October 2017, Figure S4: Averaged Rc (\(\mu\)m) from 5 to 7 January 2017 (Left: MODIS data; Mid: WDM6 Scheme; Right: Morrison Scheme), Figure S5: Averaged COD from 5 to 7 January 2017 (Left: MODIS data; Mid: WDM6 Scheme; Right: Morrison Scheme), Figure S6: Averaged CLWP (g m\(^{-2}\)) from 5 to 7 January 2017 (Left: NCEP data; Mid: WDM6 Scheme; Right: Morrison Scheme), Table S1: Comparison of observed and average Rc, COD and CLWP with CCN values of 250 cm\(^{-3}\) simulated by Morrison and WDM6 scheme from 5 to 7 January 2017.

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