Anonymous Referee #1 (Review comments in regular; response in bold.)

This paper discusses the aerosol cloud life time effect for the diurnal cycle of Stratocumulus clouds over the ARM SGP site, and comparing the results by a cloud resolving model with those of a GCM. They find that entrainment related evaporation can dominate the autoconversion reduction with increasing aerosol concentration, a mechanism not typically observed in GCMs. In general, I find this paper well written, understandable and novel, so I recommend publication in ACP after some questions are addressed.

Particularly, I am worried about the 50 to 100m horizontal resolution of the CRM, and the 30+ m in the vertical; in most LES intercomparisons of SCu, a much higher resolution is used, particularly to resolve the sharp interface of the SCu top entrainment. Do the authors have a good feel for how well their CRM is converged?

Answer: We used two horizontal resolutions for the CRM, 50 m and 100 km, not 100m. The response of LWP to increased aerosol number does change signs when we change dx=50 m to dx=100 km but not dx=100 m.

To test the convergence of the model, ideally one would increase the resolution of the CRM to see if the results still hold. Due to the limited computational resource we have, we run the cases with a decreased horizontal resolution of 100m. Both dx=50m cases and dx=100m cases show the same trend of the LWP, i.e., LWP decreases with increasing aerosol number concentrations. We agree that a resolution with dx=50m and 30+m in the vertical may be somewhat higher than most LES simulations of SCs and a smaller grid size could be better in capturing the cloud top processes and show some quantitative difference in the LWP. But we expect the trend of LWP to the increased aerosol number will be the same. As the main issue we wish to show is how basic processes left out of GCMs can lead to overestimates of the response of LWP to aerosol particle number, the exact change of LWP is less relevant here. Moreover, we never claimed that we were running the simulation at LES resolutions.

The coarse resolution case dx=100km, was a sensitivity study to mimic what occurs in single column CAM model. We chose this coarse resolution to substantially reduce the vertical movement so that the effect from the microphysics would dominate. By doing this we isolate the microphysics effect from the entrainment effect at the cloud top and demonstrate that using a reduced autoconversion rate in the CRM also increases the LWP when the aerosol number concentrations are increased, which is similar to the response in the GCM.

The other major question that I have is about the case that they chose. It is a complicated SCu case, with a strong diurnal cycle, and an a-typical qt profile. So why this case, and do you feel it is representative for cloud life time effects across the globe? Perhaps a slightly less generic title would help to lower the expectations here.

Answer: One of the targeted goals of our funded project was to use the relatively less frequently used IOPs from the SGP site. Thus we chose to use the forcing data derived from the Midlatitude Continental Convective Clouds Experiment (MC3E) which was the most recently conducted experiment near the ARM Southern Great Plains (SGP) site. For this study, May 27th, 2011, was selected because middle and

high clouds were absent during a low cloud period observed near noon. We agree that such a case may not be representative enough for SCs across the globe. But the finding (i.e., the absence of cloud-top entrainment effect in GCMs may lead to an overestimated LWP for increased aerosol numbers) should be applicable to SCs with dry air above the cloud top across the globe. To make the title less generic, we change the title from "Why do GCMs overestimate the aerosol cloud lifetime effect? A comparison of CAM5 and a CRM" to "Why do GCMs overestimate the aerosol cloud lifetime effect? A case study comparing CAM5 and a CRM".

Other points:

1) It is not very clear from your introduction that you are talking about Stratocumulus, and probably of the kind that barely precipitates. I would make that more clear in the introduction.

Answer: Done.

2) How does your work compare with the DYCOMS results, and the papers by Andy Ackerman et al? (e.g., Nature, 2005 and MWR, 2009)

Answer: Our case is similar to some extend to the DYCOMS-II case in Ackerman et al. (2004) but with even less drizzling. This makes the increased entrainment effect even more dominant than the decreased drizzling effect in our case and explains why we only see decreased LWP with increasing aerosol concentrations. We now mentioned this in the discussion in the revised manuscript.

3) P2, 1 14: Make sure to name your models, and to expand properly **Answer: Done. We now gave the full name in the abstract.**

4) P3: I am missing a description of your boundary layer scheme here. This is likely a crucial part of information for the CRM entrainment (or lack thereof).

Answer: Whereas CAM has a shallow convection scheme to parameterize the subgrid transport of heat, moisture, momentum and tracers by asymmetric turbulence within the PBL and a separate moist turbulence scheme for vertical transport by symmetric turbulence (Park and Bretherton 2009), in the CRM, subgrid-scale (turbulent) processes are parameterized using a scheme based on Klemp and Wilhelmson (1978) and Soong and Ogura (1980). The effects of both dry and moist processes on the generation of subgrid-scale kinetic energy have been incorporated. We added this information in the revised manuscript.

5) Since your resolution is on the lower side for the cloud top: What is your advection scheme in GCE?

Answer: GCE uses a positive definite advection scheme for scalar variables (Smolarkiewicz and Grabowski 1990).

6) Also, I have to ask: Is the fact that you are using a bulk micro physics scheme an issue here?

Answer: We used a 2-moment bulk microphysics scheme which predicts both number and mass mixing ratios of cloud droplets. This scheme has the capability to simulate the

effect of aerosol indirect effects even though a bin microphysics may follow some of the details more thoroughly.

7) P5, 10: The linear decrease in aerosol means that you have a decreased CCN concentration in the Boundary Layer of about 5%, if my math is correct. Why make that change?

Answer:

Yes, there is a small decrease of CCN with height in the Bounday Layer. This decrease followed the model's default set-up (the aerosol profile is fixed). We agree that the aerosol number should be (well mixed within PBL. But the small decrease assumed in the model should have a very minor impact and will not change the model behavior.

8) P7, 1: Could you plot the cloud cover as well? The dynamics may very well change as a function of aerosol (or model), for instance moving between cumulus and stratocumulus here.

Answer: We checked the cloud coverage. The change is negligible when we increase the aerosol number and could not be distinguished any change visually from a height-time plot. All clouds are stratus clouds without any cumulus cloud. In the CAM's microphysics scheme for the rain budget in warm clouds, the source terms of rain include autoconversion and accretion of cloud droplets and the only sink term is the evaporation term of rain. The relatively large decrease of surface precipitation when the aerosol number is increased from 250 to 500 cm⁻³ is a combination of decreased autoconversion/accretion and increased evaporation of rain. When the aerosol number is increased from 250 to 500 cm⁻³, the sum of autoconversion/accretion decreases. Meanwhile since there is less rain falling through the unsaturated sub-cloud layers, the final fraction of rain which can survive evaporation also decreases. The relatively large decrease of surface precipitation is peculiar to the aerosol numbers and environmental conditions. We added a statement to this effect in the manuscript.

9) P7, 1 18: To mitigate concerns about this particular case, it could be nice to quickly look at a second one as well. But at the very least, a bit more discussion about the dynamics of this case would be appreciated. (e.g., is it decoupled? What is the w2 profile? How much precipitation do you observe as a function of height).

Answer: We were not sure whether you were asking about the cases associated with number concentrations or the single day we report in the paper.

For number concentration: We checked all cases with different aerosol number concentrations and they have similar structures as showed in Fig. 2b. Fig. 2b actually already presents the results from two cases 250 cm⁻³ (dash-dotted curves) and 1000 cm⁻³ (solid curves). But the curves are overlapped. An enlarged portion to distinguish them is presented in Figure S3.

Below is a figure showing the profiles of potential temperature, specific humidity, cloud water mixing ratio, rain mixing ratio and w^2 . From the profiles of θ and q_t , we can see the clouds are not decoupled at 14:00 and 15:00. The clouds at 13:00 are a little more

complicated. The abrupt decrease of w^2 at 1.3 km and the more smooth change of cloud water q_c and larger q_t , at this height suggests the clouds above 1.3 km above may not be fully coupled to the sub-cloud layer.

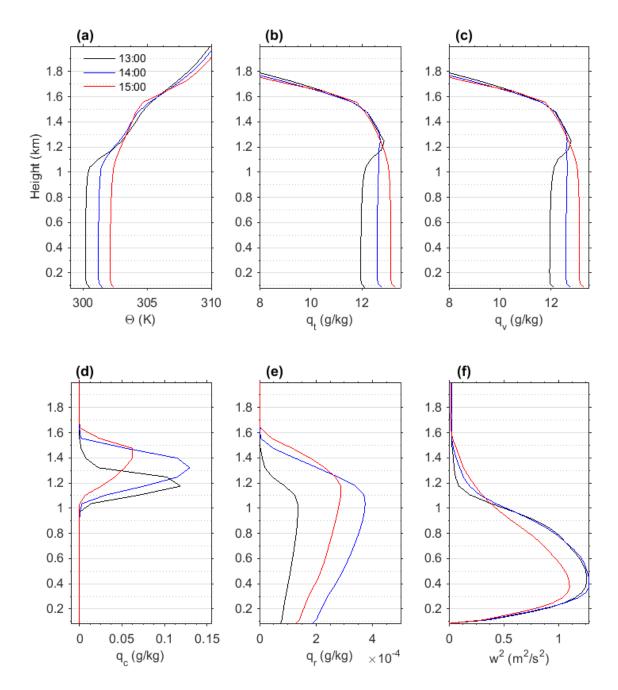


Figure 1 (a) Domain averaged potential temperatures (θ), (b) total water specific humidity (q_t), (c) specific humidity (q_v), (d) cloud water content (q_c), (e) rain mixing ratio (q_r) and (f) square of w at three times (13:00, 14:00 and 15:00) from the GCE case with surface aerosol numbers equal to 1000 cm^{-3} .

10) Why do the observations in Figure 1 show no diurnal cycle?

Answer: The clouds are modulated by advected water vapor and heat fluxes. The fluxes are showed in Fig. S1.

11) P8, 1 22: You state that some differences are likely because of details in the microphysical model. I understand that you cannot get those perfectly identical, but did you do a parameter study to get a feel of this sensitivity?

Answer: Since aerosol number affects cloud droplet number and thus directly affects the autoconversion rate, we tested the behavior the autoconversion rate to the increasing aerosol number in the two models. As showed in Figure 2 below, autoconversion rates are functions of in-cloud cloud mass mixing ratio and number mixing ratio in both models. Compared to CAM's scheme, autoconversion rates from GCE overall have a smaller dependence on cloud droplet number but larger dependence on cloud mass mixing ratio. We extracted the two pairs of in-cloud droplet number/mass mixing ratios ([26 cm⁻³, 0.167 g/kg] and [122 cm⁻³, 0.293 g/kg]) from the center layer of clouds at 11:30 hour from the two CAM cases in which the surface aerosol number increasing from 250 cm⁻³ to 1000 cm⁻³. The autoconversion rate from the Khairoutdinov and Kogan [2000] scheme used in CAM decreases from 1.86e-9 to 4.67e-10 kg/kg/s. When we applied the GCE's scheme to these two pairs of data the autoconversion rate only decreases from 1.57e-9 to 1.48e-9 kg/kg/s.

We added following after the above referenced sentence:

"Since the autoconversion rate is directly affected by the aerosol number, we used an offline model to compare the autoconversion rates from the GCE and those from the Khairoutdinov and Kogan [2000] scheme used in CAM. The results are shown in Fig. S4. Compared to CAM's scheme, autoconversion rates from the GCE are less sensitive to the droplet number concentrations when the number concentrations are less than $100 \, \mathrm{cm^{-3}}$ and the cloud mass mixing ratio is above $0.1 \, \mathrm{g \, kg^{-1}}$. When the cloud number concentrations are larger than $200 \, \mathrm{cm^{-3}}$, the autoconversion rates from GCE have a larger dependence on the number concentrations than those from the CAM scheme. However, they have a larger dependence on cloud mass mixing ratio than those from the CAM model. So increasing aerosol number tends to decrease the autoconversion rate more in CAM than in GCE. As an example, we extracted the two pairs of incloud droplet number concentrations and mass mixing ratios ([26 $\, \mathrm{cm^{-3}}$, 0.167 g kg⁻¹] and [122 $\, \mathrm{cm^{-3}}$, 0.293 g kg⁻¹]) from the center layer of clouds at the 11:30 hour from the two CAM cases in which the surface aerosol number increased from 250 $\, \mathrm{cm^{-3}}$ to 1000 $\, \mathrm{cm^{-3}}$. When applying CAM's scheme to these two pairs of data, the autoconversion rate decreases from 1.86×10^{-9} to $4.67 \times 10^{-10} \, \mathrm{kg \, kg^{-1} \, s^{-1}}$. In GCE's scheme, the autoconversion rate only decreases from 1.57×10^{-9} to $1.48 \times 10^{-9} \, \mathrm{kg \, kg^{-1} \, s^{-1}}$."

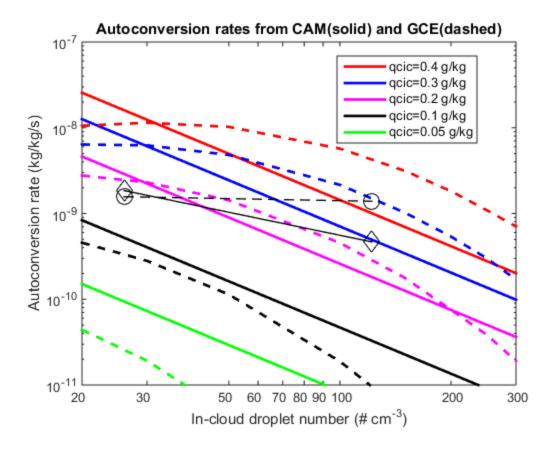


Figure 2. Autoconversion rates from the Khairoutdinov and Kogan [2000] scheme used in CAM (solid curves) and from the stochastic collection equation solutions used in GCE (dashed curves) as functions of in-cloud cloud mass mixing ratio and number mixing ratio. An air density of 1.0 kg/m3 is used. The two pairs of diamond and circle points are autoconversion rates from the two different schemes (diamond: CAM, circle: GCE) using simulated in-cloud droplet number/mass mixing ratios ([26 cm⁻³, 0.167 g/kg] and [122 cm⁻³, 0.293 g/kg]) which are extracted from the center layer of clouds at the 11:30 hour from the two CAM cases with surface aerosol number equal to 250 cm⁻³ and 1000 cm⁻³, respectively.

12) P9, 121: "..50m to 100km: ::" Should be meter, (I hope)

Answer: It is 100 km. We choose this coarse resolution on purpose to substantially reduce the vertical movement within the CRM so that the effect from the microphysics would dominate. By doing this we isolated the microphysics effect from the entrainment effect at the cloud top and demonstrated that the reduced autoconversion in the CRM would also increase the LWP when the aerosol number concentrations are increased, which is similar to that in the GCM.

Reference

Klemp, J. B., and R. B. Wilhelmson, 1978: The simulation of three-dimensional convective storm dynamics. J. Atmos. Sci.,35, 1070–1096.

Smolarkiewicz, P. K., and W. W. Grabowski, 1990: The multidimensional positive advection transport algorithm: Nonoscillatory option. J. Comput. Phys., 86, 355–375.

Soong, S.-T., and Y. Ogura, 1980: Response of tradewind cumuli to large-scale processes. J. Atmos. Sci., 37, 2035–2050.

Referee #2 Ghan (Review comments in regular; response in bold.)

This manuscript presents a clear comparison between two very different models driven by the same boundary conditions but yielding very different results. The analysis clearly reveals the causes of the differences. The work has important implications for global estimates of aerosol effects on clouds.

Lines 29-30. The word "show" is used twice in this sentence. I suggest instead "Observations of ship tracks show that the liquid water path (LWP) in marine boundary-layer clouds can either increase or decrease with increasing aerosol particles::"

Answer: Done.

Page 3. It's worth describing the subgrid treatment of cloud microphysics in CAM5: size distribution and subgrid variability.

Answer: Done. We added following in the revised manuscript:

"In-cloud cloud water variability within a GCM grid is considered and represented by an explicit gamma distribution based on observed cloud optical depth variability in marine boundary layer clouds. The subgrid in-cloud water mixing (q_c'') follows a gamma distribution $P(q_c'') = \frac{q_c''^{\nu-1}\alpha^{\nu}}{\Gamma(\nu)} \exp(-\alpha q_c'')$, where $\alpha = 1/q_c'$, q_c' is mean in-cloud mixing raito and ν is chosen to be 1 for simplicity in the model. This subgrid variability function is used to derive factors which can then be applied to calculate microphysical process rates using only the mean in-cloud mixing ratios."

Page 4. Does the GCE also use saturation adjustment? How does the dependence of autoconversion on droplet number compare with the KK scheme?

Answer: The version of GCE used in this study also uses saturation adjustment. Water vapor above saturation is removed at the nucleation step.

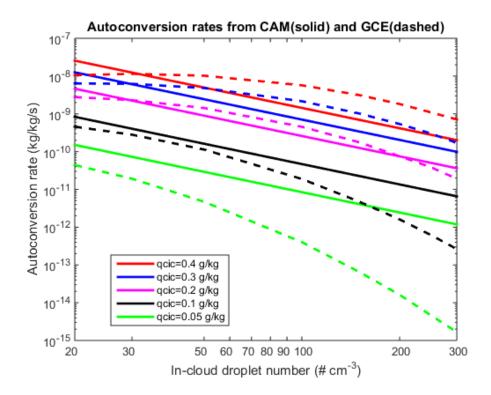


Figure 1. Autoconversion rates from Khairoutdinov and Kogan [2000] scheme used in CAM (solid curves) and stochastic collection equation solutions used in GCE (dashed curves) as a function of in-cloud cloud mass mixing ratio and number mixing ratio. An air density of 1.0 kg/m3 is used.

Figure 1 shows autoconversion rates from CAM's Khairoutdinov and Kogan [2000] scheme and the GCE. In both models, autoconversion rates are functions of in-cloud cloud mass mixing ratio (q_c) and number concentration (N_d). Compared to CAM's scheme, autoconversion rates from GCE are less sensitive to the change of N_d when N_d is less than 100 cm⁻³ and q_c is above 0.1 g/kg. When N_d is larger than 200 cm⁻³ and q_c is above 0.1 g/kg, the autoconversion rates from GCE have a larger dependence on N_d than CAM's scheme. The autoconversion rates from GCE also show larger dependence on the cloud mass mixing ratio as manifested by the wider vertical range of the curves.

Page 6, line 19. Start new paragraph here.

Answer: Done.

Page 6, line 30. Start new paragraph here.

Answer: Done.

Page 7, second paragraph. Please make clear which model is being discussed.

Answer: Done.

Page 8, lines 22-24. "This is likely due to the fact that the two models use different cloud droplet activation schemes as well as schemes to parameterize the autoconversion and accretion processes" Please demonstrate this with offline results.

Answer: Since aerosol number affects cloud droplet number and thus directly affects the autoconversion rate, we tested the response of autoconversion rate to increasing aerosol number in the two models. We extracted the two pairs of in-cloud droplet number/mass mixing ratios ([26 cm⁻³, 0.167 g/kg] and [122 cm⁻³, 0.293 g/kg]) from the center layer of clouds at the 11:30 hour from the two CAM cases in which the surface aerosol number increased from 250 cm⁻³ to 1000 cm⁻³. The autoconversion rate from Khairoutdinov and Kogan [2000] scheme used in CAM decreases from 1.86e-9 to 4.67e-10 kg/kg/s. While applying GCE's scheme to these two pairs of data the autoconversion rate only decreases from 1.57e-9 to 1.48e-9 kg/kg/s. The two pairs of autoconversion rates are added to figure 1 to form a new figure 2 as showed below. We added this figure as Fig.S4 in the supplementary material.

We added following after the above referenced sentence:

"Since the autoconversion rate is directly affected by the aerosol number, we compared autoconversion rates from GCE and Khairoutdinov and Kogan [2000] scheme used in CAM offline. The results are shown in Fig. S4. Compared to CAM's scheme, autoconversion rates from GCE less sensitive to the number concentrations when the number concentrations are less than 100 cm⁻³ and the cloud mass mixing ratio is above 0.1 g kg⁻¹. When the cloud number concentrations are larger than 200 cm⁻³, the autoconversion rates from GCE have a larger dependence on the number concentrations than the CAM scheme. However, they have a larger dependence on cloud mass mixing ratio than those from the CAM model. So increasing aerosol number tends to decrease the autoconversion rate more in CAM than in GCE. As an example, we extracted the two pairs of in-cloud droplet number concentrations and mass mixing ratios ([26 cm⁻³, 0.167 g kg⁻¹] and [122 cm⁻³, 0.293 g kg⁻¹]) from the center layer of clouds at the 11:30 hour from the two CAM cases in which the surface aerosol number increased from 250 cm⁻³ to 1000 cm⁻³. When applying CAM's scheme to these two pairs of data, the autoconversion rate

decreases from 1.86×10^{-9} to 4.67×10^{-10} kg kg⁻¹ s⁻¹. In GCE's scheme, the autoconversion rate only decreases from 1.57×10^{-9} to 1.48×10^{-9} kg kg⁻¹ s⁻¹. "

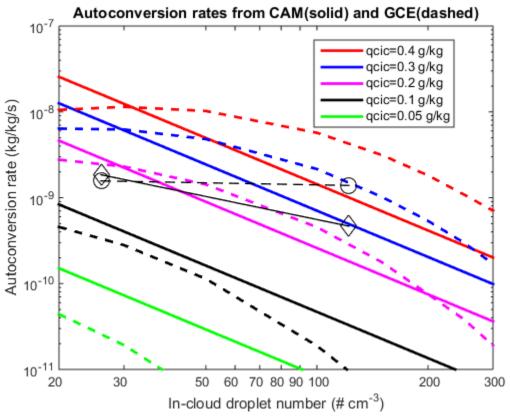


Figure 2. Same as Figure 1 except with 2 added pairs of autoconversion rates from Khairoutdinov and Kogan [2000] scheme used in CAM (diamond) and stochastic collection equation solutions used in GCE (circles). The two pairs of in-cloud droplet number/mass mixing ratio are [26 cm⁻³, 0.167 g/kg] and [122 cm⁻³, 0.293 g/kg] which are extracted from the center layer of clouds at the 11:30 hour from the two CAM cases with surface aerosol number of 250 cm⁻³ and 1000 cm⁻³ respectively.

Page 9, line 21. Insert "horizontal" before "grid".

Answer: Done.

Page 11, line 3. New paragraph.

Answer: Done.

Page 11, lines 10-13. I believe Chris Bretherton tried to implement a treatment of this mechanism in CAM but did not get the desired result. The code for that mechanism might even be in CAM5. I recommend contacting him about that.

Answer: We contacted with Chris Bretherton and cited the related work.

Why do GCMs overestimate the aerosol cloud lifetime effect? A case study comparing

comparison of CAM5 and a CRM

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Abstract

Observation-based studies have shown that the aerosol cloud lifetime effect or the increase of cloud liquid water path (LWP) with increased aerosol loading may have been overestimated in climate models. Here, we simulate shallow warm clouds on 05/27/2011 at the Southern Great Plains (SGP) measurement site established by Department of Energy's Atmospheric Radiation Measurement (ARM) Program using a single column version of a global climate model (Community Atmosphere Model or CAM5.3) and a cloud resolving model (CRM). The LWP simulated by CAM increases substantially with aerosol loading while that in the CRM does not. The increase of LWP in CAM is caused by a large decrease of the autoconversion rate when cloud droplet number increases. In the CRM, the autoconversion rate is also reduced, but this is offset or even outweighed by the increased evaporation of cloud droplets near cloud top, resulting in an overall decrease in LWP. Our results suggest that climate models need to include the dependence of cloud top growth and the evaporation/condensation process on cloud droplet number concentrations.

1. Introduction

Traditionally aerosols have been thought to lengthen cloud lifetime (Albrecht, 1989) by increasing droplet number and reducing droplet size thereby delaying and reducing the formation of rain in clouds. These longer lived clouds would then increase cloud cover and reflect more sunlight. Yet observational evidence for these lifetime effects is limited and contradictory (Boucher et al. 2013). Observations of ship tracks show that marine boundary-layer clouds polluted by aerosol particles show that the liquid water path (LWP) in marine boundary-layer clouds can either increase or decrease with increasing aerosol particles depending on factors like mesoscale cloud cellular structures, dryness of the free troposphere and boundary layer depth (Christensen and Stephens 2011; Chen et al., 2012, 2015). Results

from large-eddy simulations (LES) and cloud resolving models (CRM) show the response of cloud water to aerosols is complicated by competing effects like reduced precipitation formation efficiency in clouds and enhanced evaporation at cloud top or in the downdraft regions of cloud edges (Ackerman et al. 2004; Xue and Feingold, 2006; Tao et al., 2012). Since CRMs and LES models resolve clouds, have more complete physics and depend less on subgrid parameterizations than general circulations models (GCMs), they are often used together with field measurements to evaluate and improve parameterizations of clouds and radiation used in climate models. Several previous studies have compared single column models, which are essentially an isolated column of a GCM, and cloud resolving models (Moncreiff et al. 1997; Ghan et al., 2000; Xu et al., 2002; Xie et al., 2002; Xie et al., 2005). Lee and Penner (2010) extended these types of comparisons to the response of the two models (CAM and a CRM) to increases in aerosols in thin non-precipitating marine stratocumulus. Both models found that LWP increased but the effect from increased condensation dominated in the CRM while the effect from decreased autoconversion dominated in CAM. Wang et al. (2012) used satellite observations of the precipitation frequency susceptibility together with model simulations to constrain cloud lifetime effects in warm marine clouds simulated in GCMs. They show that GCMs tend to overestimate the precipitation frequency susceptibility of marine clouds. Since the LWP increase as a result of increased cloud condensation nuclei concentrations is highly correlated with precipitation frequency susceptibility in climate models, they surmise that the LWP increase is too high and show that this overestimation could be "fixed" by reducing the dependence of the autoconversion rate on cloud droplet number in the models.

In this study, we simulated continental shallow warm clouds with a very small precipitation rate (< 0.1 mm day⁻¹) observed on 05/27/2011 at the Southern Great Plains (SGP) measurement site established by Department of Energy's Atmospheric Radiation Measurement (ARM) Program using the single column version of a global climate model (CAM, version 5.3) and a cloud resolving model and explored plausible causes for the differences in the response of these two models to increases in aerosols. Here we specifically identify that the cloud top growth and turbulence mixing parameterizations within CAM require improvement, rather than only the autoconversion rate. Section 2 describes the models and set-up. Section 3 presents results followed by conclusions and a discussion in section 4.

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2. Description of models and set-up

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We used the Goddard Cumulus Ensemble model (GCE) with recent improvements (Tao et al. 2014) and the single column version of Community Atmosphere Model (CAM, version 5.3) which is the atmospheric component of the Community Earth System Model (CESM, version 1.2.2). Readers are referred to Neale et al. (2012) for more model details of CAM. Here we briefly summarize the two most critical parameterizations for warm stratus clouds in CAM: cloud microphysics and cloud macrophysics. The cloud microphysics (version MG1.5) is a two-moment scheme (Morrison et al. 2005, Morrison and Gettelman 2008) which predicts the number concentrations and mixing ratios of cloud droplets. The source term for the cloud droplets in warm clouds only includes the activation of cloud condensation nuclei while the sink terms include the instantaneous evaporation of falling cloud droplets into the clear portions of grids beneath clouds, autoconversion of cloud droplets to form rain, and accretion of cloud droplets by rain. The first two sink terms (instantaneous evaporation of falling cloud droplets and autoconversion) depend on the aerosol number concentration since the terminal falling speed of cloud droplets is related to cloud droplet size and the autoconversion rate is inversely proportional to cloud droplet number ($\sim N_c^{-1.79}$ where N_c is the in-cloud cloud droplet number). The last sink term (accretion) does not depend on the cloud droplet number (Khairoutdinov and Kogan 2000). In-cloud cloud water variability within a GCM grid is based on observed cloud optical depth variability in marine boundary layer clouds. Thus, the sub-grid in-cloud water mixing (q'') follows a gamma distribution $P(q_c'') = \frac{q_c''^{\nu-1}\alpha^{\nu}}{\Gamma(\nu)} exp(-\alpha \ q_c''), \ \underline{\text{where}} \ \alpha = 1/q_c', \ \underline{q_c' \ \text{is mean in-cloud mixing raito and}} \ \underline{\nu} \ \underline{\text{is}}$ chosen to be 1 for simplicity. This sub-grid variability function is used to derive factors which can then be applied to calculate microphysical process rates using only the mean incloud mixing ratios. The conversion of water vapor to cloud condensate is computed by the cloud macrophysics parameterization which also predicts the cloud fraction in each grid as well as the horizontal and vertical overlapping structures of clouds. Following Smith (1990), the liquid fraction of stratus clouds in CAM5 is derived from an assumed triangular distribution of total relative humidity (i.e. the sum of water vapor and liquid cloud water). The net conversion rate of water vapor to stratus condensate is diagnosed using saturation equilibrium conditions: (1) the RH over the water within the liquid stratus is always 100%, and (2) no liquid stratus droplets exist in the clear portion of the grid.

The Goddard Cumulus Ensemble model (GCE) is a CRM that has been developed and improved at the NASA Goddard Space Flight Center (GSFC). Its development and main features were published in Tao and Simpson (1993) and Tao et al. (2003) and recent improvements and applications were presented in (Tao et al. 2014). The GCE model used in the present paper uses the double moment version of the Colorado State University Regional Atmospheric Modeling System (RAMS) bulk microphysics scheme (Saleeby and Cotton, 2004) which assumes a gamma-shaped particle size distribution for three species of liquid (small and large cloud droplets and rain). The small cloud droplets range from 2 to 40 microns in diameter, and the large cloud droplets range from 40 to 80 microns. Collection of cloud droplets is simulated using stochastic collection equation solutions, facilitated by binemulating look-up tables. A positive definite advection scheme is used for scalar variables (Smolarkiewicz and Grabowski 1990). Sub-grid-scale (turbulent) processes are parameterized using a scheme based on Klemp and Wilhelmson (1978) and Soong and Ogura (1980). The effects of both dry and moist processes on the generation of sub-grid-scale kinetic energy have been incorporated. Readers are referred to Lee et al. (2009) and Tao et al. (2014) for more detailed descriptions of the model physics.

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CAM has 30 vertical layers and a variable vertical resolution which depends on the surface pressure and the vertical temperature profile. In the case studied in this paper the vertical resolution is roughly 100 meters near the surface and stretches to about 300 m at 2 km decreasing to ~1 km at 10 km. The time step is 20 minutes. GCE has 128 grids in the two horizontal directions and 144 vertical layers. The horizontal resolution is 50 m, so the domain size is 6.4 km × 6.4 km. GCE also uses a stretched vertical resolution that varies from about 30 m near the surface to about 90 m at 2 km and further to ~200 m at 10 km. The time step of the GCE model is 1 second. Both models use the same initial conditions (surface pressure/temperature, vertical temperature/water vapor/wind profiles), boundary conditions (surface sensible/latent heat fluxes, surface pressure/temperature). Advective tendencies of temperature and moisture (both vertically and horizontally) are specified based on an objective variational analysis approach (Xie et al. 2014) fit to the Midlatitude Continental Convective Clouds Experiment (MC3E) campaign observations which were conducted from April to June 2011 near the DOE ARM Southern Great Plains (SGP) site. The analyzed advective tendencies cover the period from April 22nd to June 21th, 2011. Middle to deep convective clouds were observed in most cloudy days. For this study, May 27th, 2011, was selected because middle and high clouds were absent during a low cloud period observed near noon. The vertical wind/temperature/moisture/cloud fraction profiles, surface

latent/sensible heat fluxes, and advective tendencies of temperature and moisture are shown in Fig S1. Low clouds occurred from ~1 km to ~ 2 km near the top of the boundary layer and were strongly modulated by the advective tendencies of temperature and moisture. Positive moisture flux and negative temperature flux were observed during the growing stage of the clouds while negative moisture flux and positive temperature flux were observed during the decaying stage. Both models are initialized at 00:00 local time and run for 18 hours.

To study the effect of aerosols on clouds, we scaled the aerosol vertical profiles in both models by increasing the surface aerosol number concentrations from 250 cm⁻³ to 4000 cm⁻³. GCE uses a prescribed aerosol profile which decreases linearly from its surface concentration to 100 cm⁻³ at an altitude of 14 km and above. The activation of aerosols to cloud droplets is based on the grid resolved vertical updraft velocity, temperature, and aerosol number and size from a look-up table constructed from results of a Lagrangian parcel model (Saleeby and Cotton, 2004). For CAM, we extracted the averaged aerosol profile in May at this location from a 5-year run of CAM5 using the MAM3 aerosol module and scaled the aerosol profile based on the surface aerosol number concentrations (see Fig. S2 for profiles of aerosol number concentrations used in the two models). The activation of aerosols into cloud droplets in CAM is diagnosed as a function of the modeled subgrid-scale updraft velocity and aerosol compositions/sizes/numbers (Abdul-Razzak and Ghan 2000). Even though we set the total surface aerosol number concentrations the same in the two models, the aerosol composition, size, and number at cloud level, and the nucleation schemes are inherently different. However, since this paper focuses on a sensitivity study which is aimed at revealing the different cloud physical representations in the two models that lead to *opposite* responses of the simulated LWP to increasing aerosol number concentrations that cover a wide range (250 cm⁻³ to 4000 cm⁻³) rather than quantifying the changes of the LWP, these differences are not critical to the conclusions of the paper. To better isolate differences in the aerosol indirect effect in the two models, we also turned off the aerosol direct radiative effect.

3. Results

Figure 1a shows the observed cloud fractions from the early morning to the late afternoon on May 27th, 2011 at the SGP site, while Figures 1b and 1c show the simulated mean cloud water content from the two models assuming a surface aerosol number concentration of 500 cm⁻³. Compared to the observations, the simulated clouds from both models begin later in the day and have a smaller vertical coverage. But the models compare relatively well to each other which suggests that differences between the models and the observations may largely

be caused by the possible errors/uncertainties associated with the derived initial conditions or advective tendencies. Nevertheless, we can see that the GCE model captures the observed growth of the clouds with height while CAM does not. A detailed analysis of the GCE (next paragraph) shows that the clouds could be loosely classified as stratocumulus which occur near the top of the planetary boundary layer (PBL) and are mainly driven by long wave radiative cooling offset by short wave radiative heating. This is corroborated by CAM's result which shows all simulated clouds are stratus clouds and no convective clouds are able to form above the PBL. The advective tendencies of heat and moisture also strongly modulate the clouds. For example, the positive moisture tendency before 14:00 hours leads to slightly larger in-cloud water vapor mixing ratio than that below the clouds (more details will be presented in the discussion of Figure 2).

Figure 1d and 1e show the domain averaged liquid water path (LWP) from the two models for five different surface aerosol number concentrations (250, 500, 1000, 2000 and 4000 cm⁻³). Both models underestimate the LWP during the day, similar to their underestimation of cloud cover. GCE shows relatively small changes in the LWP when using different surface aerosol numbers. The LWP slightly increases with the increasing aerosol number before ~14:00 but starts to decrease with the increasing aerosol number when the clouds start to decay after around 14:00. On the other hand, the LWP from CAM increases substantially and consistently with increasing aerosol number and matches the observed LWP better when the surface aerosol number is equal to 4000 cm⁻³. As noted earlier, due to uncertainties associated with the derived forcing data as well as uncertainties in the models, this should not be interpreted as proof that CAM represents the physics better.

Figure 1f and 1g show the precipitation rates from the two models. The precipitation rate from CAM consistently decreases with increasing aerosol number and is nearly suppressed after 13:00. The change is most prominent when the aerosol number is increased from 250 to 500 cm⁻³. This result is due to a combination of decreased autoconversion/accretion and increased evaporation of rain. When the aerosol number is increased from 250 to 500 cm⁻³, the sum of autoconversion/accretion decreases. Meanwhile since there is less rain falling through the unsaturated sub-cloud layers, the final fraction of rain which can survive evaporation also decreases. The relatively large decrease of surface precipitation is peculiar to the aerosol numbers and environmental conditions simulated here. The precipitation rates from GCE are overall very small with maximum values less than 0.08 mm day⁻¹. The change in precipitation for GCE with increasing aerosol numbers is a little more complex. During the

growing phase of the clouds, as in CAM, the precipitation rate decreases. But during the decaying phase, the precipitation rate actually increases even though the LWP decreases.

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Figures 2a-2c show the domain averaged potential temperatures (θ), total water specific humidity (q_t) and cloud water content (q_c) at three times (13:00, 14:00 and 15:00) from the two CRM cases with surface aerosol numbers equal to 250 cm⁻³ (dash-dotted curves) and 1000 cm⁻³ (solid curves), respectively. q_t is the sum of q_c, rain and water vapor mixing ratios, which is an invariant within the PBL for stable non-precipitating well-mixed stratocumulus. θ and q_t from the two cases almost overlap except near the cloud top at 14:00 and 15:00. Fig. 2a shows the growth of the PBL. At 13:00 the clouds do not completely reside within the PBL as the top of the PBL is at about 1.2 km which is lower than the cloud top height (~1.5 km) shown in Fig. 2c. Fig. 2b shows that q_t in the top half of the cloud (from ~1.2-1.5 km) is larger than q_t in the bottom half of clouds (from ~1-1.2 km) and q_t below the clouds at 13:00. This suggests that the top half of the clouds are not fully coupled with the surface and the cloud water in the top half of the clouds is strongly affected by the horizontally advected positive moisture flux. At 14:00 and 15:00, the advected moisture flux becomes negative and the PBL is high enough that the clouds reside fully within the top of the PBL and possess the characteristics of well-mixed stratocumulus. The domain averaged long-wave cooling rate at the cloud top height is about 2 K hr⁻¹ and is offset by a short-wave heating of about 0.5 K hr⁻¹. Fig. 2c shows that the cloud top is a little higher for the higher aerosol case, but the maximum values of q_c are smaller. A closer look at θ in Fig. 2a also shows that the top of the PBL which is near 1.5 km is higher and colder in the higher aerosol number case. These differences of q_c and θ between the two cases are clearer in an enlarged portion of Fig 2a and 2b shown in Fig. S3. The potential temperature in the sub-cloud layer at 14:00 and 15:00 is also slightly higher (about 0.005 K) for higher aerosols. Figs. 2d to 2i show the time-averaged profiles of q_c and the net result of condensation and evaporation (Conden-Evap) during two 1-hour intervals (Fig. 2d-f for 13:00 to 14:00 and Fig. 2g-i for 14:00 to 15:00) representing the growing and decaying phases of the cloud, respectively. Figures 2e and 2h show that a net evaporation occurs just below the cloud base and near the cloud top. The largest net condensation is located near the cloud base. The most obvious change between the growing phase and decaying phase of the cloud is the increased evaporation near the cloud top, especially for the high aerosol number case (see the changes from blue curve to the red curve at around 1.5 km from Fig. 2e and Fig. 2h). Choosing (Conden – Evap)/ q_c as a measure of the inverse of the characteristic evaporation time of

cloud droplets, Figures 2f and 2i show that it increases substantially from 300 hr⁻¹ to about 600 hr⁻¹ (an evaporation time of ~6 seconds) near the cloud top for the higher aerosol number case.

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Figure 3 shows the LWP and the column integrated LWP source and sink terms from the low and high aerosol cases (250 and 1000 cm⁻³). The source term for LWP only includes the net condensation term (Conden - Evap) while the loss terms include autoconversion and accretion. Since CAM includes a separate autoconversion and accretion terms while GCE does not, we combined autoconversion and accretion as one term (Auto+Accre) for easier comparison. As shown in Fig. 1, when we increase the aerosol numbers from 250 to 1000 cm⁻¹ ³, the LWP increase is relatively small in GCE and substantially larger in CAM. Both models show decreased Auto+Accre which acts to increase the LWP. This is expected as increased aerosol numbers increase the cloud droplet number which decreases the autoconversion rate. But CAM shows much larger changes, especially before 13:00 hours. This is likely due to the fact that the two models use different cloud droplet activation schemes as well as schemes to parameterize the autoconversion and accretion processes. Since the autoconversion rate is directly affected by the aerosol number, we used an offline model to compare the autoconversion rates from the GCE and those from the Khairoutdinov and Kogan [2000] scheme used in CAM. The results are shown in Fig. S4. Compared to CAM's scheme, autoconversion rates from the GCE are less sensitive to the droplet number concentrations when the number concentrations are less than 100 cm⁻³ and the cloud mass mixing ratio is above 0.1 g kg⁻¹. When the cloud number concentrations are larger than 200 cm⁻³, the autoconversion rates from GCE have a larger dependence on the number concentrations than those from the CAM scheme. However, they have a larger dependence on cloud mass mixing ratio than those from the CAM model. So increasing aerosol number tends to decrease the autoconversion rate more in CAM than in GCE. As an example, we extracted the two pairs of in-cloud droplet number concentrations and mass mixing ratios ([26 cm⁻³, 0.167 g kg⁻¹] and [122 cm⁻³, 0.293 g kg⁻¹]) from the center layer of clouds at the 11:30 hour from the two CAM cases in which the surface aerosol number increased from 250 cm⁻³ to 1000 cm⁻³. When applying CAM's scheme to these two pairs of data, the autoconversion rate decreases from 1.86×10^{-9} to 4.67×10^{-10} kg kg⁻¹ s⁻¹. In GCE's scheme, the autoconversion rate only decreases from 1.57×10^{-9} to 1.48×10^{-9} kg kg⁻¹ s⁻¹. –Moreover, in GCE, the decreased autoconversion is largely offset or even outweighed by increased evaporation. As shown in Fig. 2e and 2h the increased evaporation occurs near cloud top. The increased evaporation near the cloud top and the higher PBL suggests that higher aerosol number concentrations

lead to smaller cloud droplet sizes and enhanced evaporation at the cloud top which can then decrease the temperature slope near the cloud top and promote the sinking of entrained air into the cloud layer, a point made previously by Bretherton et al. (2007). This evaporationentrainment feedback mechanism was also observed in small cumulus clouds (Small et al. 2009). Before ~14:00, the effect from the decreased autoconversion rates outweighs the effect from increased evaporation so that the LWP shows a slight increase. But as the cloud after ~14:00, the PBL keeps growing and the starts enhanced evaporation/entrainment rates accelerate the decaying process. Thus the LWP decreases faster and eventually a smaller LWP results over the decaying period for the high aerosol case. In the CAM model, the change of the net condensation term (Conden – Evap) is smaller than that in the CRM model. Since the simulated cloud top remains unchanged between 12:00 and 15:00 hours, the drying effect seen in the CRM due to enhanced entrainment of overlying dry air is not present. This is likely due to the fact that the moist turbulence scheme in CAM does not depend on the cloud droplet number/size and the condensation and evaporation in the CAM's macrophysics scheme is not linked to the cloud droplet number or size. Even though the instantaneous evaporation of falling cloud droplets into the clear portions of grids beneath clouds in the microphysics scheme is related to the cloud droplet number, it is about one order of magnitude smaller than the net condensation term in the macrophysics scheme. Consequently the net condensation and evaporation is less sensitive to the change in aerosol number and the effect from the decreased autoconversion rate dominates the condensate loss, leading to an increase of the LWP.

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To confirm that the effect from enhanced entrainment at the cloud top is the critical reason for the reduced LWP change in GCE, we ran a sensitivity test to reduce the cloud top mixing by increasing the horizontal grid spacing from 50 m to 100 km. With this larger grid spacing, we greatly reduced the overshooting at the cloud top by reducing the maximum vertical speed in the updrafts from meters per second to a few centimeters per second. As a result, the enhanced entrainment effect was reduced and the microphysical effect from the reduced autoconversion rate dominated. Figure 4 shows that the LWP from GCE decreases by about 5% for the dx=50 m case while it increases by about 12% for the dx=100 km case when the surface aerosol number is increased from 250 cm⁻³ to 4000 cm⁻³. We also ran two more tests to explore whether the LWP sensitivity in CAM could match that in the GCE. In the default set-up of CAM, the autoconversion rate is inversely proportional to cloud droplet number ($\sim N_c^{-1.79}$ where N_c is the in-cloud cloud droplet number). We ran two cases, auto06

and auto00, each with a reduced dependence of the autoconversion rate on the cloud droplet number. In case auto06, the autoconversion rate is proportional to $N_c^{-0.60}$ and in case auto00, the autoconversion rate does not depend on the cloud droplet number. The autoconversion rate is scaled in both cases to produce the same rate as that from the default case at a droplet number concentration of 100 cm⁻³. As shown in Fig. 4, the LWP from the default case is more than doubled when the surface aerosol number is increased from 250 cm⁻³ to 4000 cm⁻³ while the LWP from auto06 only increases by ~50% and the LWP from case auto00 remains almost unchanged. These results suggest that the dependence of the autoconversion rate on the cloud droplet number can play a determining role on the simulated LWP consistent with the findings of precipitation frequency susceptibility in Wang et al. (2012). However, this adjustment is unable to simulate decreases in LWP seen in the GCE model.

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4. Conclusion and Discussion

We simulated shallow warm clouds on May 27th, 2011 at the DOE ARM SGP site with a cloud resolving model (Goddard Cumulus Ensemble model) and a single column model (CAM) using the same initial/boundary conditions and advected moisture/heat tendencies derived from the MC3E campaign data. The liquid water path (LWP) simulated by CAM shows a large dependence on the aerosol loading and is more than doubled when the surface aerosol number is increased from 250 cm⁻³ to 4000 cm⁻³ while the LWP simulated by the CRM decreases by ~5%. The high sensitivity of LWP on aerosol loading in CAM can be reduced by reducing the dependence of the autoconversion rate on the cloud droplet number concentration, but is unable to reproduce the decrease in LWP seen in the CRM. Whereas Wang et al. (2012) concluded that this term in GCM models can be tuned to fit observations of the precipitation frequency susceptibility, we find that the poor representation of entrainment and droplet evaporation in CAM model may be the fundamental cause of differences with the more complete CRM. While in the CRM a reduced autoconversion rate is also observed with increased aerosol loading, it is offset or even outweighed by the increased evaporation of cloud droplets near the cloud top. The increased evaporation cools the cloud top, reduces the temperature lapse rate and thus increases the entrainment of drier air above the cloud top and accelerates the decaying process of the clouds. Reduced LWP through enhanced entrainment with increased aerosol number has also been reported in previous literature using large eddy simulations (e.g., Ackerman et al. 2004, Bretherton et al. 2007, Seifert et al. 2015). To some extent our case is similar to the DYCOM-II case studied

in Ackerman et al. (2004) with low humidity above the cloud top. Our case has even less drizzling and this makes the increased entrainment effect even more dominant than the decreased drizzling effect which explains why we only see decreased LWP with increasing aerosol concentrations.

One unique aspect of the present paper is that the response of the LWP over the lifetime of the cloud is negative in the CRM while it is positive in the CAM model for the same forcing conditions. One critical deficiency of CAM for this case is that the effect from increased mixing of drier air from above the cloud layer through enhanced entrainment caused by increased aerosol numbers is missing. First, CAM is not able to simulate the growth of the cloud top due to its coarse vertical resolution. However, even if the CAM vertical resolution were high enough to capture the growth of the cloud top, since the moist turbulence scheme and the evaporation of cloud condensate in the cloud macrophysics parameterization at the cloud top are not related to the cloud droplet number, aerosol number will not have a direct impact on the cloud top mixing or the LWP. Some effort has been made to address this issue in CAM. Jones (2013) implemented a droplet sedimentation-entrainment feedback scheme in CAM. Yet a mixture of the cloud macrophysics and the MG microphysics still prevent clouds from responding to droplet number changes by thinning or thickening as demonstrated by other LES simulations.

Our CRM model results <u>also</u> demonstrate that the relative importance of the decreased autoconversion rate effect and the enhanced entrainment effect from increased aerosol numbers can change based on environmental conditions as manifested in different stages during the cloud lifecycle. Thus, one may need to distinguish the cloud stage when studying the aerosol lifetime effect either with a model or from observations.

5. Acknowledgements

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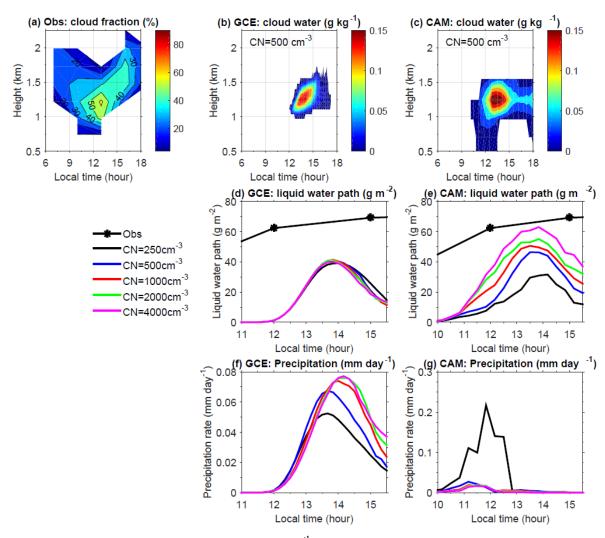


Figure 1. Observed cloud fractions on May 27th, 2011 at the SGP site (a); domain averaged cloud water content from the GCE model (b) and the single column version of CAM (c) for the case assuming a surface aerosol number of 500 cm⁻³; liquid water path and surface precipitation rates from GCE (d, f) and CAM (e, g) with varying surface aerosol number concentrations.

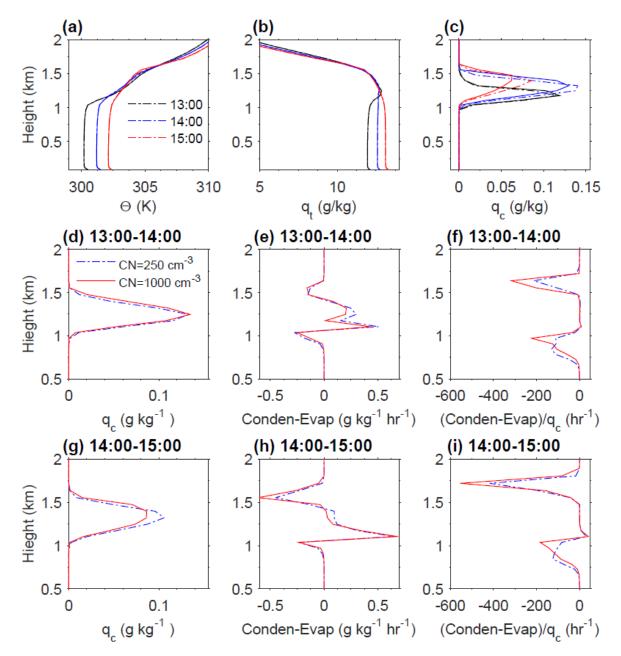


Figure 2. (a-c) Domain averaged potential temperatures (θ), total water specific humidity (q_t) and cloud water content (q_c) at three times (13:00, 14:00 and 15:00) from two GCE cases with surface aerosol numbers equal to 250 cm⁻³ (dash-dotted curves) and 1000 cm⁻³ (solid curves). (d-f) Averaged profiles of q_c , net results of condensation and evaporation (Conden-Evap), and (Conden-Evap)/ q_c for the 1-hour interval from 13:00 to 14:00 from the two CRM cases with surface aerosol numbers equal to 250 cm⁻³ (blue dash-dotted curves) and 1000 cm⁻³ (solid red curves). (g-i) Same as (d-f) except for the 1-hour interval from 14:00 to 15:00.

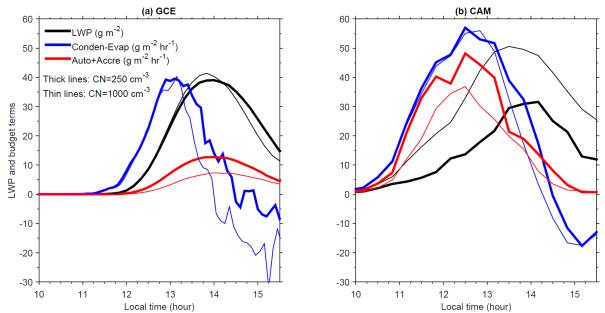


Figure 3. LWP and the column integrated LWP source and sink terms from the case with surface aerosol number concentration equal to 250 cm⁻³ (thick lines) and 1000 cm⁻³ (thin lines) for (a) GCE and (b) CAM.

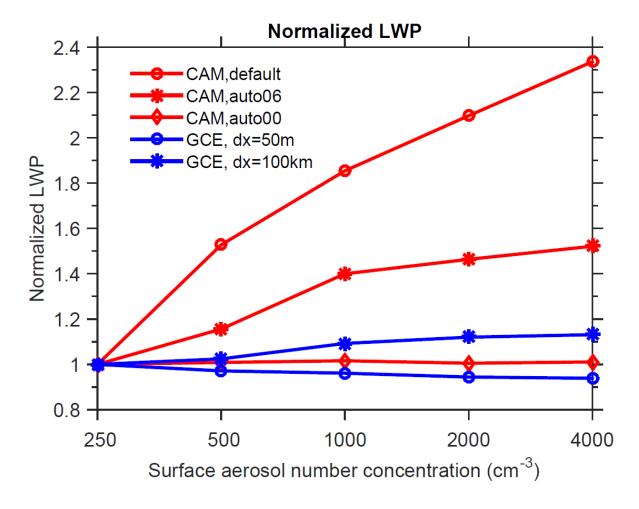


Figure 4. Normalized LWP as a function of surface aerosol concentration in CAM (red curves) and GCE (blue curves). A case for CAM using an autoconversion rate proportional to $N_d^{-0.6}$ (CAM, auto06) as well as a case in which autoconversion is independent of N_d (CAM, auto00) is shown. The GCE model was run with a horizontal grid resolution of 50 m (default case) and 100 km.