Modeling the impact of aerosols on tropical overshooting thunderstorms and stratospheric water vapor

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Overshooting deep convection plays an important role in regulating the water vapor content of the tropical tropopause layer and is also an important mechanism for transporting water vapor into the lower stratosphere (LS). The aim of this study is to examine the effect of aerosols as cloud condensation nuclei (CCN) on the water vapor content of the LS via single isolated overshooting thunderstorms. The development of a severe Hector thunderstorm in northern Australia observed during the Stratospheric–Climate Links with Emphasis on the Upper Troposphere and Lower Stratosphere/Aerosol and Chemical Transport in Tropical Convection (SCOUT-O3/ACTIVE) campaign is simulated using a three-dimensional nonhydrostatic convective cloud model with a double-moment bulk microphysics scheme. The results show that the ice hydrometeors account for over 50% of the total condensate mass, indicating that ice processes play an important role in regulating the structure of thunderstorms in the tropics. A large number of ice particles occurring in the LS are not formed in situ but are transported upward by convective overshooting with subsequent mixing. Sensitivity tests show that the increase in cloud droplet numbers induced by increasing CCN concentrations would increase the number concentrations of the ice crystals transported to the LS, which had the effect of reducing the sizes and fall speeds of the ice crystal, thereby causing a moistening of the LS by sublimation of the injected ice particles. This result suggests that aerosols in the boundary layer can affect stratospheric water vapor via overshooting deep convection.


1. Introduction

[2] Changes in water vapor in the upper troposphere and lower stratosphere (UT/LS) play an important role in modulating the Earth’s climate since it is the most powerful greenhouse gas in the atmosphere. An increase of about 1% in water vapor per year in the stratosphere was observed over the past 50 years [Oltmans et al., 2000; Rosenlof et al., 2001]. Determining what affects the stratospheric water vapor content is important for better understanding the greenhouse effect and global climate change.

[3] Water vapor in the stratosphere has two main sources. One is transport of water vapor from the troposphere via deep convection to the tropical tropopause layer (TTL), from there to the stratosphere by the Brewer-Dobson circulation. The other is the oxidation of methane, which occurs mostly in the upper stratosphere. The tropics are the main source regions of water vapor in the Earth’s atmosphere. Deep convection is occurring very frequently in these regions, and some of the deepest convection can penetrate up to the cold point tropopause (CPT) and have significant overshoots. In recent years, the contribution of overshooting deep convection to the water vapor content of the TTL and lower stratosphere (LS) has attracted considerable attentions [Romps and Kuang, 2009; Toon et al., 2010]. Previous studies suggest that overshooting dehydrates the stratosphere caused by the irreversible mixing of colder, drier air into the ambient environment [Danielsen, 1982, 1993; Sherwood and Dessler, 2000, 2001]. More recently, however, measurements [Nielsen et al., 2007; Corti et al., 2008; de Reus et al., 2009; Khaykin et al., 2009; Iwasaki et al., 2010] and model simulations [Chaboureau et al., 2007; Chemel et al., 2009; Grosvenor et al., 2007; Liu et al., 2010; Wang, 2003] suggest that overshooting convection has rather a hydrating than dehydrating effect by sublimation of the injected ice particles above the tropopause. Furthermore, Liu et al. [2010] and Grosvenor [2010] have made quantitative estimates of the impact of overshooting convection on the stratospheric water vapor content. Their results showed that the mass of water vapor transported into the stratosphere was associated with the severity of the overshoot. Hassim and Lane [2010] reported the importance of TTL relative humidity in governing the stratospheric moisture, and showed that overshooting tended to hydrate (dehydrate) the stratosphere when the TTL was already subsaturated (supersaturated).
[4] Tropical deep convection mainly occurs over three regions, namely South America, Central Africa, and the Maritime Continent, which includes the Indonesian archipelago and northern Australia [Chen et al., 2009]. During the Australian premonsoon and monsoon-break periods between November and February, severe thunderstorms, known colloquially as “Hector,” occur almost daily over northern Australia, particularly in the area of Darwin, as a result of the strong convergence of warm, moist sea and land breezes [Crook, 2001]. Owing to its unique location, the Darwin area has hosted a number of field campaigns devoted to tropical deep convection, including the Island Thunderstorm Experiment [Keenan et al., 1989], the Maritime Continent Thunderstorm Experiment [Keenan et al., 2000], the Stratospheric-Climate Links with Emphasis on the Upper Troposphere and Lower Stratosphere/Aerosol and Chemical Transport in Tropical Convection (SCOUT-O3/ACTIVE) [Vaughan et al., 2008], and many others. SCOUT-O3/ACTIVE was held in November–December 2005 and was aimed at determining the impact of deep convection on the composition of the UTLS region and its relative importance compared with large-scale ascent. The Hector thunderstorms were a particular focus of the SCOUT-O3/ACTIVE. One of the objectives of the SCOUT-O3/ACTIVE was to investigate the aerosol effects on deep convection.

[5] Atmospheric aerosols, formed from natural and anthropogenic processes [Zhang, 2010], as cloud condensation nuclei (CCN), can markedly affect the development of clouds both microphysically and dynamically [Andreae et al., 2004; Carrió et al., 2007; Li et al., 2008, 2009; Khain et al., 2005, 2008; Khain and Lynn, 2009; May et al., 2009; Sherwood, 2002; van den Heever et al., 2006; Zhang et al., 2007]. These will potentially alter the ability of convective clouds to transport of water and vapor into the TTL and LS. However, there have been relatively few studies of the impact of aerosols on stratospheric water vapor through its effect on tropical deep convection. Recently, Grosvenor et al. [2007] employed the UK Met Office LEM (Large Eddy Model) with bulk microphysics scheme to simulate the development of a storm complex over the Bauru area in Brazil under different aerosol conditions. Their results showed that an increase in the CCN number increased the ice water content of the TTL and the water vapor content of the LS. It is noted that the CCN number in Grosvenor et al.’s work actually refers to the activated droplets, and is assumed constant throughout the cloud’s lifetime.

[6] Darwin in northern Australia is a natural laboratory for the study of deep tropical convection responding to the variation in aerosols. There is a significant seasonal variation in aerosol characteristics in the Darwin region [Allen et al., 2008]. In the premonsoon periods, the atmosphere is relatively smoky owing to widespread biomass burning in northern Australia, resulting in a high concentration of aerosols, while in the monsoon periods, the atmosphere is back to clean maritime air owing to the cessation of the biomass burning source. The aim of this study is to determine the possible impact of severe thunderstorms that developed over the Darwin region under different aerosol conditions on the water vapor content of the LS. Two problems are addressed. One is whether aerosols affect the development of a single isolated Hector thunderstorm. The other is whether these changes in the clouds induced by the aerosols affect the stratospheric water vapor content. To address these issues, we employ a three-dimensional nonhydrostatic convective cloud model with a double-moment bulk microphysics scheme and explicit droplet activation from CCN (detailed description in section 2), in contrast to the hybrid bulk microphysics scheme employed by Grosvenor et al. [2007], which was one moment for liquid and rain and two moment for ice, snow, and graupel. Furthermore, the current work differs in that single isolated severe thunderstorms over the tropics are simulated. Although there have been several numerical studies focusing on the aerosol effects on cloud structures and precipitation of isolated thunderstorms [e.g., Cui et al., 2006; Yin and Chen, 2007; Lerach et al., 2008; Khain and Lynn, 2009], few were based on the tropics and they did not focus on the impact of aerosols on how severe tropical thunderstorms affect stratospheric water vapor.

[7] The rest of the paper is organized as follows: The model description is provided in section 2, followed by the simulation initialization in section 3. The dynamics and microphysics of the simulated thunderstorms developed under two CCN conditions are investigated in section 4, along with a change of water vapor and ice content in the LS induced by CCN. Finally, section 5 presents a summary and the conclusions.

2. Model Description

[8] The cloud model used in this study is a three-dimensional compressible nonhydrostatic cloud model with a double-moment bulk microphysics scheme [Kong et al., 1990; Hong and Fan, 1999; Chen and Xiao, 2010]. The model domain is on a standard spatially staggered mesh system. A conventional time-splitting integration technique, proposed by Klemp and Wilhelmson [1978], is used in the model. The large and small time steps are 2 s and 0.25 s, respectively. The spatial differential terms are of second-order accuracy except for the advection term that has fourth-order accuracy. All other derivatives are evaluated with second-order centered differences. The radiation boundary conditions of Klemp and Wilhelmson [1978] are used for the lateral boundaries while the top and bottom boundaries are assumed as a rigid wall. A Rayleigh friction zone is also used to absorb vertically propagating gravity waves near the top of the domain. The damping depth from model top is set to 6 km. The model includes a conventional first-order closure for subgrid turbulence and a diagnostic surface boundary layer based on Monin–Obukhov similarity theory.

[9] Using bulk microphysics parameterization, the mixing ratios and number concentrations of seven hydrometeor types are predicted in the model: cloud water, cloud ice, rain, snow, graupel, frozen drops and hail. The cloud droplets, ice crystals and raindrops are assumed to follow gamma size distribution, while other hydrometeors are represented by inverse exponential size distribution [Hong and Fan, 1999]. Corresponding microphysical processes include (1) nucleation of cloud droplets and ice crystals; (2) autoconversion of cloud water to rain, cloud ice to snow or graupel, snow to graupel, and graupel and frozen drops to hail; (3) homogeneous freezing of cloud water to cloud ice, rain to graupel or frozen drops, collisional freezing of rain to graupel or frozen drops, collisional freezing of hail to graupel or frozen drops; (4) accretion of cloud water by rain,
cloud ice, snow, graupel, frozen drops, and hail; (5) accretion of rain by graupel, frozen drops, and hail; (6) rain shed from graupel, frozen drops, and hail; (7) melting of cloud ice, snow, graupel, frozen drops, and hail; (8) accretion of cloud ice by snow, graupel, frozen drops, and hail; (9) accretion of snow by graupel, frozen drops, and hail; (10) self-accretion of cloud ice and snow; (11) condensation/evaporation of cloud water and rain; (12) deposition/sublimation of cloud ice, snow, graupel, frozen drops, and hail; and (13) riming-splintering of snow, graupel, and frozen drops. A complete description of the microphysical parameterization is given by Hong and Fan [1999] and Chen and Xiao [2010]. Only the key processes pertinent to the present work are described here.

[10] A power law activity spectrum of CCN is applied for droplet nucleation. The number of activated CCN, \( N_{CCN} \), for supersaturation with respect to water \( S_w \) (in percent) can be written as

\[
N_{CCN} = CS_w^k,
\]

where \( C \) and \( k \) are constant parameters. Owing to the lack of direct measurements of CCN concentration in the Darwin region, the coefficients were set to \( C = 100 \text{ cm}^{-3} \) and \( k = 0.462 \) for maritime conditions, or \( C = 1260 \text{ cm}^{-3} \) and \( k = 0.308 \) for continental conditions, on the basis of the work of Khain et al. [2005]. The CCN concentrations are assumed to decrease exponentially with altitude with a scale height of 2.5 km. Activation only occurs in regions of upward motion and when the supersaturation resolved on the model grid is positive. Nucleation of droplets may occur at cloud base or within the cloud. The peak supersaturation at the cloud base is diagnosed analytically [Twomey, 1959], and the potential numbers of droplets activated are given by Rogers and Yau [1989].

\[
N_{CCN} = 0.88C^{2/(k+2)}[0.07w^{3/2}]^{1/(k+2)},
\]

where \( N_{CCN} \) is in \( \text{cm}^{-3} \) and \( w \) is the vertical velocity in \( \text{cm s}^{-1} \). Within the cloud, the supersaturation resolved on the model grid is then applied for droplet nucleation. Calculation of the supersaturation during the time step is based on the approach of Morrison et al. [2005, equations (3)–(7)], but the present work neglects the impact of radiative transfer and microphysical processes associated with hail on supersaturation. In this scheme, the evolution of the supersaturation due to condensation/deposition (evaporation/sublimation), the Bergeron–Findeisen mechanism and adiabatic (vertical air motion) cooling is predicted over longer time scales (>1 s). The phase relaxation time scales associated with each hydrometeor species (cloud water, rain, cloud ice, snow, graupel and frozen drops) are given by Morrison et al. [2005, equation (4)], but the spectral shape index \( p_R \) used herein is specified a constant value, particularly, \( p_R = 0 \) for snow, graupel and frozen drops, 5 for cloud droplets, 2 for raindrops, and 1 for ice crystals [Hong and Fan, 1999]. The average radius of newly nucleated droplets or initial droplet size distribution is assumed to be a function of the supersaturation—the higher the supersaturation, the smaller the average radius, using the empirical formula and specific parameters given by Reisin et al. [1996]. This approach causes a relatively wide spectrum of droplets for maritime clouds compared to continental clouds, leading to drop collisions early in the maritime clouds.

[11] Rainwater can be initiated by autoconversion of cloud droplets to raindrops, melting of precipitating ice, or shedding of excess water drops accreted by graupel, frozen drops and hail in the wet growth regime. The autoconversion represents a key process in cloud development, in which raindrops are initiated by collision and coalescence of cloud droplets. Although various parameterizations of the autoconversion processes are available [see Li et al., 2008], it is beyond the scope of this study to examine in detail the impact of different schemes. We also note that Li et al. [2008] found a similar variation of cloud properties with CCN concentration produced by different types of autoconversion parameterization, especially when the CCN concentration was less than 5000 cm\(^{-3}\). A sensitivity test of different autoconversion parameterizations has been performed, by running the scheme proposed by Khairoutdinov and Kogan [2000], and the results are very similar to the present results except for the total mass of water and vapor in clouds. Autoconversion of cloud droplets to rain used herein is parameterized following Lin et al. [1983] except that the constant relative dispersion has been replaced by a predicted value based on the concentration of cloud droplets. The autoconversion term \( P_{RAUT} \) is expressed by

\[
P_{RAUT} = \rho_w(l_{CW} - w_0)^2 \cdot \left( 1.2 \times 10^{-4} + 1.569 \times 10^{-12} \frac{N_C}{D_0(l_{CW} - w_0)} \right)^{-1},
\]

where \( l_{CW} \) is the mixing ratio of cloud water, \( N_C (\text{cm}^{-3}) \) is the number concentration of cloud droplets, and \( D_0 \) is the relative dispersion. The \( w_0 \) is the threshold for autoconversion with a value of 2 \( \times 10^{-3} \) \( \text{g cm}^{-3} \). In this study, the relative dispersion \( D_0 \) is calculated from the predicted \( N_C \) using the relation [Grabowski, 1999]

\[
D_0 = 0.146 - 5.964 \times 10^{-2} \ln(N_C/2000).
\]

Furthermore, we add the mass of a droplet of a critical size, \( D_{w,auto} \), as an additional threshold for autoconversion following Phillips et al. [2007], except that \( D_{w,auto} \) is set equal to 28 \( \mu \text{m} \) to prevent unrealistically fast conversion for maritime clouds, in which the largest droplet is about 22 \( \mu \text{m} \) in diameter.

[12] The natural cloud ice is produced through condensation freezing and deposition nucleation, and homogeneous freezing of cloud droplets below -40°C. The number concentration of active natural ice nuclei is parameterized following Cooper [1986]. Once formed, cloud ice grows via the depositional and riming processes. Ice multiplication, or the secondary ice generation mechanism, for riming of snow and graupel/frozen drops at temperatures between -3°C and -8°C is based on the work of Hallett and Mossop [1974] and is parameterized following Hu and He [1988]. Snow can be formed by the Bergeron–Findeisen process and autoconversion of cloud ice. Frozen drops can be initiated by homogeneous or probabilistic freezing of raindrops, or collisions between rain and cloud ice or snow only when the raindrop
diameter is greater than 1 mm. If the raindrop diameter is smaller than 1 mm, a frozen raindrop is converted to graupel. Graupel may be also created via a parameterized form of the Bergeron process, or by aggregation of ice crystals and snowflakes. The autoconversion rate coefficient for cloud ice to form snow and for snow to form graupel is based on the work of Lin et al. [1983]. Graupel and frozen drops convert to hail when their diameters are greater than or equal to 5 mm via autoconversion [Hu and He, 1988].

Additionally, it should also be noted that the terms continental and maritime relate in this study to CCN distribution and concentration only.

3. Model Initialization

The numerical simulations presented here were based on a case study day of the 2005 SCOUT-O3/ACTIVE field campaign. The day chosen was 30 November. That day was
also a “golden Hector day” of the campaign [Brunner et al., 2009]. Hector that developed on that day was much taller and stronger than in other days of the campaign. This Hector produced a significant overshoot of convective turrets reaching up to 18 km into the stratosphere. Very light upper level winds caused the anvil to flow radially out from the storm with a slight northeastward drift. The CPT was situated at 17.3 km altitude during that day [de Reus et al., 2009]. According to de Reus et al. [2009], the ice crystals were observed at altitudes between 18 and 18.7 km, at potential temperatures level between 386 and 414 K, which corresponds to 0.7 to 1.4 km above the CPT. The presence of these ice particles would likely lead to a moistening of the LS. The effect of overshooting on hydrating the LS via sublimation of the injected ice particles from the troposphere has been further confirmed through numerical simulations by Chemel et al. [2009].

[15] The model was initialized on the basis of the rawinsonde sounding shown in Figure 1 taken from Darwin, on 30 November 2005 at 1430 LT. The sounding shows large water vapor availability with mixing ratios of 17 g kg$^{-1}$ near the surface and a warm cloud base with a temperature of more than 20°C at about 1.5 km, while the 0°C freezing temperature is at 5.1 km. The 380 K potential temperature level is situated at $\sim$17.3 km, which corresponds to the 87 hPa pressure level (not shown in Figure 1). The atmospheric environment has a very large convective available potential energy (CAPE) of approximately 3500 J kg$^{-1}$. The wind hodograph shows a low-level northwesterly flow with low shear, the flow above being westerly up to about 12 km and easterly aloft. This synoptic environment favors the triggering of Hector thunderstorms over the islands, just as suggested, for instance, by Crook [2001].

[16] All simulations were integrated to 4200 s using a grid size of 0.5 km horizontally and 0.3 km vertically, with cor-

**Figure 2.** Modeled radar reflectivity from the maritime CCN simulation at (a) 32 and (b) 40 min. Observations made by Bureau of Meteorology Research Centre–National Center for Atmospheric Research C-band polarimetric (BMRC/NCAR C-POL) radar at (c) 0510 and (d) 0540 UTC.
responding dimensions of 41 and 24 km for the model domain. A domain moving method, where the grids move with the center of the total hydrometeor mass, is used to keep the simulated storm within the computational domain. Convection was initiated by a warm thermal bubble of 20 km wide and 3 km deep, which was centered at 1.5 km above ground level in a horizontally homogeneous environment. The maximum thermal perturbation was 2.0 K in the center of the bubble with the heating rate decreasing according to a cos⁻² relationship with radial distance from the center. Such heating resulted in a relatively strong updraft and formation of the convective clouds. However, we note that Grosvenor et al. [2007, hereinafter G07] and Grosvenor [2010, hereinafter G10] found that the moistening potential of the stratosphere exhibited a large variation with bubble temperature perturbations, showing an increasing stratospheric water vapor content with increasing perturbations. In our simulations, with a smaller temperature perturbation and bubble width the simulated convection was relatively weak and failed to produce convective overshoots. On increasing the temperature perturbation and width, the simulated convection was considerably stronger when compared with the radar observations. In addition, similarly to G07 and G10, the stratospheric water content was increased with increasing temperature perturbation. On the basis of these sensitivity tests, we choose a maximum temperature perturbation of 2 K and bubble width of 20 km to initialize the convection. The following analysis is based on these choices. Figure 2 shows a comparison of the observational radar data with the simulated radar reflectivity which is calculated from the sum of the reflectivity for all hydrometeors except for cloud water [Hogan et al., 2006]. The characteristic structure of observed thunderstorms has been well reproduced by the simulations. The simulated radar reflectivity field generally agrees with the observations. The horizontal and vertical scales of convection in the simulations are similar to the radar observations. However, the simulated radar reflectivity values are somewhat higher than the observations, probably owing to the over prediction of graupel, frozen drops and hail (Figure 3), which is likely caused by the microphysical schemes and the initialization of the warm bubble. Overall, these choices of bubble width and temperature perturbation lead to the best comparison with the radar observations.

4. Results
4.1. Simulation of Convective Clouds Under Maritime Aerosol Conditions

[17] The dynamical and microphysical characteristics of simulated convective clouds in the maritime aerosol conditions will be first examined. Figure 4 presents the time evolution of the maximum updraft velocities in the model domain. One can see that updrafts tend to increase with

Figure 3. The mass mixing ratios (g kg⁻¹) of (a) 32 min and (b) 40 min, showing the presence of graupel/frozen drops and hail in high radar reflectivity corresponding to Figures 2a and 2b. Blue, cyan, yellow, red, and green lines represent mixing ratio contours of cloud ice, snow, graupel/frozen drops, hail, and rain, respectively. The black line with the value of 0.001 g kg⁻¹ indicates the cloud boundary, which is calculated from the sum of the mass density for all hydrometeors. Temperature contours are superimposed at 20°C intervals.

Figure 4. Time-height plots of the maximum updraft under maritime CCN conditions. Data are obtained from instantaneous model output every 1 min. Contours are in intervals of 4 m s⁻¹ starting at 4 m s⁻¹. The corresponding potential temperature (K) for significant altitude derived from the sounding data is plotted on the right axis.
height, with peak values of greater than 40 m s\(^{-1}\) appearing at approximately 12–14 km altitude. This result agrees with observation that peak updrafts in deep tropical convection are almost always above the 10 km level [e.g., May and Rajopadhya, 1999; Heymsfield et al., 2010]. The simulated maximum updraft velocity is 43.1 m s\(^{-1}\). This value seems to be reasonable on the basis of the prestorm environment shown in Figure 1. Using the value of 3500 J kg\(^{-1}\) for CAPE and the relation \(W_{\text{max}} = \sqrt{2} \times \text{CAPE}\), the theoretically maximum updraft velocity turns out to be 84 m s\(^{-1}\). When considering precipitation loading, mixing effects of ambient air and perturbed vertical pressure gradients, that value should be reduced by roughly 50% [Bluestein, 1993], giving a maximum updraft velocity of 42 m s\(^{-1}\), which is fairly consistent with the value simulated by our model. After 32 min the maximum vertical velocities decrease with time.

[10] The first updraft penetrates the TTL (hereinafter defined as the region between approximately 14 and 17.4 km in altitude) after 27 min and about 4 min later the updrafts have overshoted the CPT to reach into the LS. The updrafts within the TTL and LS persist for over 20 min and \textsim 10 min, respectively, allowing time for ice and vapor to be transported upward across the tropopause and into the stratosphere. Note that a strong convection appears to detach from the top of the main convection during the time period 37–50 min. The center of this convection is situated at the 18.3 km level, the peak updraft velocity is 16.8 m s\(^{-1}\), and the top reaches over 22 km. This strong updraft plays an important role in determining the entry of the tropospheric ice particles into the LS. According to Lane et al. [2003] and Wang [2003], when significant wind shear exists near the tropopause, a condition to favor gravity wave breaking, turbulence is generated at cloud top owing to gravity wave breaking. In this case, the vertical wind shear in the vicinity of the tropopause is relatively weak (Figure 1b), such that the new convection in the LS is not generated by the breaking of gravity waves.

[10] Overshooting is defined as clouds with their tops located higher than the CPT. The general structure and evolution of the simulated storm associated with the overshooting plume are shown in Figure 5 in detail with a series of vertical cross sections along the domain center. At 30 min, an updraft from 6 to 18 km has fully developed, pushing the convective dome (herein using a total water mixing ratio threshold of 10\(^{-3}\) g kg\(^{-1}\)) close to the CPT, while a low-level downdraft has established owing to precipitation loading. Temperature of the middle-low troposphere in cloud is higher than the environment owing to the diabatic heating resulting from the latent heat released by freezing of liquid water droplets within the updraft. Note that air parcels in the upper troposphere become progressively colder with height than the environment, and eventually form a cold trap region near the tropopause. This cold trap is likely due to cooling near the tropopause caused by a combination of adiabatic ascent and irreversible diabatic mixing of air overshooting its level of neutral buoyancy [Sherwood, 2000; Robinson and Sherwood, 2006]. Sherwood [2000] further argued that this cooling near the tropopause would drive descent above vigorous convection and lead to compensating ascent through the tropopause occurring elsewhere. An unusual circulation thus occurs in the troposphere and stratosphere. In our simulations, this stratosphere-troposphere circulation is established within several minutes after the cold trap (Figure 5b). Subsequently, a new convection occurs 1–2 km above the tropopause and develops rapidly. At 40 min a strong convection has established well in the LS, just above the main storm (Figure 5c). The associated cloud cover extends upward into the LS and the cloud top reaches 20 km altitude, whereas the main updraft and total water content below the CPT are decreasing rapidly. The vertical motion structure in the troposphere is also changed, with one inner updraft region and one outer updraft region between 10 and 16 km in altitude. The inner region is around the center of the storm, and the outer region is adjacent to the edge of the storm. The outer updrafts extend the cover of the cloud anvils, pushing more ice at a larger horizontal extent upward into the TTL even the LS. The mass and thermal exchanges between the storm and the environmental air are enhanced at the locations of the outer updrafts. At 50 min the updraft has decreased to 2 m s\(^{-1}\) leaving a weak perturbed circulation and the cloud dissipated by precipitation and mixing (Figure 5d). These results indicate that the overshooting could be an important mechanism for transporting water into the stratosphere. After 50 min rather ice water of 0.001–1 g kg\(^{-1}\) is still remained in the TTL (Figures 5e and 5f).

[20] Lin et al. [2005] found that the ratio of ice mass to liquid mass in the maritime subtropical thunderstorms is \textsim 1:1. This demonstrates that ice processes are highly important to cloud structure even in thunderstorms occurring in warm climatic regimes. In our simulations, in a time-averaged sense, the hydrometeor mass partitioning in the domain is 8.2% for cloud water, 41.5% for rainwater, 18.7% for cloud ice, 3.6% for snow, 8.6% for graupel, 12.9% for frozen drops and 6.5% for hail, so that the ice phase accounts for about 51% of the total hydrometeor mass. This percentage is very similar to that of the subtropical thunderstorm presented by Lin et al. [2005]. Note also that cloud ice accounts for about 19% of the total hydrometeor mass, contributing 40% of the total ice mass.

[21] Corti et al. [2008] investigated this event in details and found that the observed ice water content in the stratosphere was much too high to be formed in situ. He suggests that the ice particles in the stratosphere likely result from overshooting convection of the Hector. In our simulations, with the mean over the whole 41 \times 41 km domain and over the whole 70 min duration, 5.6 ppmv and 3.1 ppbv of total ice in the form of cloud ice are found at 17.4 km and 20.1 km in altitude, respectively (Figure 6). The maximum cloud ice mixing ratio reaching these two levels is 2186.8 ppmv occurring at 34 min and 6.7 ppmv at 44 min, respectively. How are these high mixing ratios of stratospheric ice particles produced? There are two potential mechanisms for cloud ice formation in the simulation: In situ nucleation and upward transportation from the cloud top. In the model, there are three generation mechanisms of ice crystals: primary nucleation, riming splintering and homogeneous freezing of cloud droplets. On the basis of the simulation conducted here, we found that these three processes occurred primarily below 14 km owing to the limitation of water vapor and temperature, as shown in Figure 7a, while depositional growth and sublimation processes of cloud ice crystals occurred in the stratosphere indeed (Figures 7b and 7c). After having excluded the in situ formation, the simulated cloud ice particles in the stratosphere
Figure 5. Vertical cross sections of total water content larger than $10^{-3}$ g kg$^{-1}$ (white), updraft greater than 2 m s$^{-1}$ (black), and the temperature field (shaded, °C) along x = 20 km under maritime CCN conditions for the time after (a) 30 min, (b) 35 min, (c) 40 min, (d) 50 min, (e) 60 min, and (f) 70 min. The red line represents the 380 K potential temperature isotherm.
are solely transported upward by convective overshooting with subsequent mixing. This is similar to the conclusion made by Corti et al. [2008] based on observations of this cloud. The upward transporting ice crystal across the tropopause is found to occur during the time period 30–34 min and 38–50 min. Through calculating the vertical flux of ice crystal concentrations, we found that the contribution induced by the second stage was over 75% of the total, so that the second stage was very important for ice production in the LS. After 48 min, evaporation of cloud ice is almost the only microphysical process occurring in the LS (Figure 7c).

Figure 8 shows the time evolution of the mean water vapor mixing ratios at each level above 14 km altitude. One can see that a significant dry layer is present between 16 and 18 km in altitude at the early stage of the simulations. After 27 min, the water vapor mixing ratio within the TTL is rapidly increased, indicating a moistening of the TTL due to deep convection. Meanwhile, the water vapor in the LS is also progressively enhanced. At the end of the simulation, the total mass of water vapor is ~4140 tons (1000 kg) for the TTL and 1366 tons for the LS, increased by about 128% and 5%, respectively, compared to the simulation start. By the end of the simulation, accumulated total mass of water vapor in the LS is 92990 tons, during the 40 min integration period covering the overshoot, the total mass of water vapor is 53618 tons, thereby resulting in an extra 14247 tons of water vapor entering the LS owing to the overshoot (Table 1). These results clearly demonstrate that ice particles injected by overshooting have a moistening effect on the LS.

During the time period covering the overshoot, the total mass of cloud ice entering the stratosphere is 56104 tons with a corresponding increase rate of 23 tons s$^{-1}$. Note that rather total water is still left in the cloud at the end of the simulation (Figure 5f). The amount of cloud ice that remains in the stratosphere is about 2500 tons. This suggests that the...
moistening of the LS is potentially increased further given more time.

4.2. Simulation of Convective Clouds Under Continental Conditions

While clouds in both the maritime and the continental cases began to form after 7 min of simulations, the clouds in the latter case produced much more cloud water than in the former. The maximum cloud droplet concentration in the maritime case is only 82 cm\(^{-3}\) while 814 cm\(^{-3}\) were produced in the continental case. Cloud water exists primarily below the melting level (5.1 km) in the maritime case (Figure 9a), whereas in the continental case, the cloud water exists as high as 10 km, at temperatures near –40°C. The core of the cloud water is at 5–6 km or 0°C to –5°C (Figure 9b) with the maximum LWC of 7.5 g m\(^{-3}\). In comparison, only 4.7 g m\(^{-3}\) was obtained in the maritime case.

Higher droplet concentrations in the continental case lead to stronger competition for available water vapor during droplet growth via diffusion and therefore restrict the size of cloud droplets. As a consequence, droplet growth by the collision–coalescence process is less efficient in the continental case than in the maritime case, resulting in a time delay (~6 min) of raindrop formation in the continental case (Figures 9e and 9d). The initial raindrop formation is found in the continental case at a much higher altitude than in the maritime case (5.1 versus 2.4 km). Although the accretion of cloud water is the dominant growth mechanism for rain during the first 30 min in both cases, accounting for about 90% of the rain production, in the case of high CCN concentrations, rain develops more slowly and less effectively as compared to the case of low CCN concentrations, resulting in much lower rainwater content. After 30 min, nearly all of the rainwater is below the 0°C level in both cases, which indicates that the melting of ice hydrometeors is the main source of the rain production. The largest source of the rainwater is the melting of frozen drops in the maritime case, and melting of graupel in the continental case. Melting of hail is the second largest source in both cases. The maximum rainwater content is 14.5 g m\(^{-3}\) in the maritime case versus 12.8 g m\(^{-3}\) in the continental case, indicating that the increase in aerosols in the continental thunderstorm produces more cloud water but much less rain, as compared to the maritime thunderstorm. Correspondingly, the accumulated precipitation at the ground is also reduced with increasing CCN concentrations (Table 1).

The continental clouds have more ice crystals and less snow compared to the maritime clouds (Figures 9e–9h). Most of cloud ice is located generally above 11 km, at temperatures below –40°C, and are formed by homogeneous freezing of cloud droplets. The maximum concentration reaches 60 cm\(^{-3}\) in the continental case and 30 cm\(^{-3}\) in the maritime case, both occurring at 33 min. The mass of ice crystals in the continental case is also slightly higher than that in the maritime case, with a maximum in the domain averaged values of 0.984 g m\(^{-3}\) versus 0.978 g m\(^{-3}\). The higher ice crystal concentration in the continental case leads to stronger competition for available water vapor during depositional growth and therefore restricts the size of ice crystals. As a result, the snow process is less efficient in the continental case.

In the model, freezing raindrops can convert into graupel or frozen drops depending on the raindrop diameter \(D_{fr}\). The destination particle is then classified as graupel when \(D_{fr} < 1\) mm, and as frozen drop when \(D_{fr} > 1\) mm. The fraction of small raindrops with \(D_{fr} < 1\) mm increases with increasing CCN concentrations. Subsequently, the concentration of graupel particles in the continental case (with a maximum of ~60 L\(^{-1}\)) is significantly higher than that in the maritime case (6 L\(^{-1}\)). The graupel mass content in the continental case is also significantly higher than that in the maritime case, with a maximum of 12.0 g m\(^{-3}\) versus 1.0 g m\(^{-3}\). But the maritime clouds have many more frozen drops (with the maximum of 900 m\(^{-3}\) in the concentration and 6.6 g m\(^{-3}\) in mass content) compared to the continental clouds (760 m\(^{-3}\) and 4.4 g m\(^{-3}\)).

The high CCN concentration leads to more hails in the continental case than in the maritime case (Figures 9m and 9n). The maximum values are 20 m\(^{-3}\) for number concentration and 8.8 g m\(^{-3}\) for mass content in the continental case, while only 10 m\(^{-3}\) and 4.7 g m\(^{-3}\) appears in the maritime case. The increase in the hail is mainly due to the increase in

### Table 1. Cloud Properties Under the Maritime (Case-M) and Continental (Case-C) CCN Conditions

<table>
<thead>
<tr>
<th>Case-M</th>
<th>Case-C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum updraft in the cloud (m s(^{-1}))</td>
<td>43.1</td>
</tr>
<tr>
<td>Maximum updraft in the LS (m s(^{-1}))</td>
<td>16.8</td>
</tr>
<tr>
<td>Accumulated total mass of condensate (KT)</td>
<td>54515</td>
</tr>
<tr>
<td>Ratio of ice mass to liquid mass</td>
<td>~5:5</td>
</tr>
<tr>
<td>Total increase in stratospheric ice caused by overshoot (tons)</td>
<td>56104</td>
</tr>
<tr>
<td>Accumulated total mass of stratospheric vapor (tons)</td>
<td>92990</td>
</tr>
<tr>
<td>Total increase in stratospheric vapor caused by overshoot (tons)</td>
<td>14247</td>
</tr>
<tr>
<td>Accumulated total mass of liquid precipitation (KT)</td>
<td>1686</td>
</tr>
<tr>
<td>Accumulated total mass of solid precipitation (KT)</td>
<td>97</td>
</tr>
</tbody>
</table>

*Maximum updraft is given by an instantaneous value. The accumulated total mass is the overall amount produced throughout the simulation.
Figure 9. Time-height plots of domain-averaged hydrometeor mass density (g m$^{-3}$) under (a, c, e, g, i, k, m) maritime and (b, d, f, h, j, l, n) continental CCN conditions.
autoconversion of graupel and accretion of cloud water by hail, as shown in Figures 9i and 9j).

[29] From Figure 10 and Table 1, it can be found that in the high CCN concentration case (the continental case), total condensate mass for the entire thunderstorm system is increased by 4.2%, as compared to that in the low CCN concentration case (the maritime case). This result indicates that the total condensate mass in tropical thunderstorms increases with an increase in the CCN concentration. Furthermore, the partitioning of hydrometeors in thunderstorms is also related to the CCN concentration. As shown in Figure 10, the continental thunderstorms have more cloud water, cloud ice, graupel and hail mass, but less rainwater and frozen drop mass than in the maritime case. Overall, ice hydrometeors account for about 57% of the total hydrometeor mass, showing that the fraction of ice phase in the thunderstorms increases with increasing CCN concentrations.

[30] Figure 11 shows the time evolution of the maximum updraft velocity at each level during each 1 min interval. Compared to the maritime case, the clouds that developed in the high CCN concentration case have larger updraft velocities before 28 min, occurring at the middle levels. As shown in Figures 9a and 9b, most of the cloud water develops before 30 min in the simulations. But the continental case produces more latent heat during condensation and freezing because of the higher droplet concentration and the higher levels that the droplets reach, leading to the increase in the updraft velocity. In the maritime case, low CCN concentrations lead to the formation of raindrops at lower levels. In the absence of vertical wind shear (Figure 1b), the effects of the latent heat of condensation on the cloud dynamics are offset by the precipitation loading mainly in the form of rain, resulting in smaller vertical velocities in the maritime case than in the continental case. After 28 min, however, the maximum updraft velocities in the maritime case in the upper levels of the cloud are higher than in the continental case which produced a peak updraft of 37.2 m s$^{-1}$. On the basis of the aforementioned microphysical analysis, one can see that the continental clouds have more precipitation mass in the upper levels during the mature stage of thunderstorm, especially the graupel (Figures 9i and 9j) and hail (Figures 9m and 9n). The loading effect of precipitation mass suppresses the upward motion and causes the storm to have a weaker updraft as compared to the case of low maritime CCN concentrations. Correspondingly, a weaker cold trap region is formed in the continental case, thus resulting in a weaker descent above the cloud top and a weaker convection in the LS with the maximum of 14.5 m s$^{-1}$. This result indicates that increasing CCN concentration weakens the intensity of overshooting tropical thunderstorms.

4.3. Sensitivity of Ice and Water Vapor Content in the LS to CCN Concentrations

[31] Considering that the cold point tropopause was situated at 17.3 km altitude for this event and the vertical grid size used in the simulations, the LS boundary in our work is defined at 17.4 km. Figure 12 shows the stratospheric mean changes in total ice and water vapor content between the
continental and maritime cases. One can see that the increase in CCN leads to increases in both the ice mass and number concentrations and vapor content in the LS. For water vapor, maximum increments in the domain and time averaged values of 4.4 ppbv were found in the LS occurring at 17.7 km altitude. This change is directly associated with homogeneous freezing of cloud droplets and subsequent transporting upward by overshooting. The increase in CCN concentration cause a large increase in the number of small ice particles formed owing to droplet homogeneous freezing process, since the more numerous small droplets form more ice particles when they reach the homogeneous freezing zone and then freeze. This subsequently results in more ice particles being advected up into the LS, so that the ice crystal number concentrations within the LS are eventually increased. Corresponding to the significant increase in the number concentration of ice crystals, which would have the effect of reducing the sizes and fall speeds of the ice crystal, the vapor mass is increased subsequently by sublimation of the injected cloud ice particles.

To better understand the effect of the increasing CCN concentrations on ice and water vapor in the LS, Figure 13 presents the time evolution of the difference between the continental and maritime cases in accumulated total mass. The result shows that the rapid increase in ice water content occurs after convective overshooting especially for the time period 30–34 min when the thunderstorm enters the mature stage. This increase is attributed to a large number of the injected ice crystals by strong updrafts. Within the following several minutes, the increasing effect is reduced. As mentioned before, during 35–37 min the vertical motion above the cloud top is dominated by descent, thus some ice mass in the LS are transported downward across the tropopause and back into the troposphere. After 38 min, the ice mass in the LS begins increasing slowly when the stratosphere-troposphere convection develops above the cloud top. However, the increase in ice water mass during the vigorous stratosphere-troposphere convection is smaller, when compared to the time period 30–34 min. After 45 min, the ice water mass in the LS is progressively increased although there is little vertical transporting of ice crystals across the tropopause when the stratosphere-troposphere convection decayed. This increase in the ice mass is wholly caused by less precipitation loss of ice by lower sedimentation. In the continental case, the size and fall speed of the ice crystals is smaller than that in the maritime case. Overall, the mean diameter and fall speed of the ice crystal in the LS for the continental clouds has been reduced by 40% and 22%, respectively. Lower fall speeds lead to less precipitation loss of ice by sedimentation and allow more ice to remain in the LS. Overall, after 50 min when there is no vertical motion near the tropopause, the increase in ice mass is 5570 tons, accounting for 70% of the total increase, so that the sedimentation effect dominates the ice mass in the LS. Furthermore, slow sedimentation of the ice leads to increased evaporation upon mixing with the surrounding air, thereby causing an increase in the stratospheric water vapor content.

By the end of the simulation, the accumulated increases caused by increasing CCN in the LS are 8089 tons for cloud ice and 96 tons for water vapor, with a corresponding increase of 14.4% and 0.1%, respectively. At the end of the simulation, the amount of cloud ice that remains in the stratosphere is 2774 tons in the continental case. That value is higher than in the maritime case. This suggests that the stratospheric water vapor would be potentially increased further given more time.

5. Summary and Conclusions

In this paper we investigate the effects of aerosols acting as CCN on tropical overshooting thunderstorms and
stratospheric water vapor using a three-dimensional non-hydrostatic convective cloud model with a double-moment bulk microphysics scheme and explicit droplet activation. A severe Hector thunderstorm that developed on 30 November 2005 during the SCOUT-O3/ACTIVE campaign was simulated under maritime and continental aerosol scenarios.

[35] The results show that ice-phase hydrometeors account for over 50% of the total hydrometeor mass of the tropical thunderstorms. This is consistent with the subtropical thunderstorms simulated by Lin et al. [2005] and further demonstrates that ice processes are highly important in determining cloud structure in thunderstorms occurring in warm climate regimes. Ice crystals are highly important and contribute about 19% of the total hydrometeor mass in this severe tropical thunderstorm. It is also found that the cloud ice particles in the LS are not formed in situ but are transported upward by convective overshoots with subsequent mixing occurring. The penetration of convective overshoots has a moistening effect on the LS by sublimation of the injected ice particles.

[36] It is shown that aerosols significantly influence the cloud microphysics and dynamics of the simulated tropical thunderstorms. An increase in the CCN concentration leads to more cloud water, cloud ice, graupel and hail, but less rainwater and frozen drops. Total cloud condensate mass is enhanced as well. Moreover, the partitioning to ice-phase hydrometeors in the storms increases with an increase in the CCN concentration. With more ice crystals being transported upward into the LS, which would have the effect of reducing the sizes and fall speeds of the ice crystal, the stratospheric water vapor content is increased by sublimation of the injected ice particles. This result suggests that aerosols in the boundary layer do affect the entry of water vapor in the LS, in agreement with the work of Sherwood [2002].

[37] Note that the simulated overshooting convection in the high CCN concentration case was found to be weakened somewhat owing to the loading effect of increased precipitation mass at the middle and upper levels, when compared to the low CCN concentration case. In this work, however, microphysical effects induced by aerosol increases dominate, resulting in a moistening effect on the stratosphere. In contrast, a reduced moistening effect might be produced if dynamical effects play a dominant role. If this was the case then this would suggest that the dependence of the water vapor content of the LS on aerosol concentrations might be nonlinear. Connolly et al. [2006] and Li et al. [2008] found nonmonotonic change of the convection intensity with aerosols, because of the complicated coupling between cloud microphysics and dynamics. The present work only investigated one case and two aerosol scenarios, more cases and sensitivity tests are needed to determine whether aerosols increase or decrease the stratospheric water vapor.

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References


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