Effect of aerosol on the electrical structure of thunderstorms

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Abstract: Numerical simulations are performed to investigate the effect of aerosol on microphysical and electrification in thunderstorm clouds. A two-dimensional (2-D) cumulus model with electrification scheme including non-inductive and inductive charge separation is used. The concentration of aerosol particles with distribution fitted by superimposing three log-normal distribution function rises from 50 to 10000 cm^{-3}.

The results show that the charge structure in thundercloud keeps as a triple with aerosol concentration increasing. When aerosol concentration is changed from 50 to 1000 cm^{-3}, a stronger formation of cloud droplet, graupel and ice crystal result in increasing charge separation via non-inductive and inductive mechanism. However, in the range of 1000-3000 cm^{-3}, the decrease of ice crystal caused by vapor competition leads to the reduce of upper positive charge, while the enhance of graupel and cloud droplet result in the contribution of inductive charge to the middle negative charge region and lower positive charge region increased with greater aerosol concentration. At very high aerosol concentration (above 3000 cm^{-3}), the magnitude of charge which remains steady in thundercloud is insensitive to aerosol concentration.

Introduction

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Many studies have been devoted to space charge distributions in thunderstorms, that is closely related to the characteristics of lightning discharge (Carey and Rutledge, 1998; Coleman *et al*., 2003; Qie *et al*., 2005; Tan *et al*., 2006, 2012, 2014). A host of observations of soundings of the electric field demonstrate that the complex charge structure usually including four to ten charge layers in thunderstorms (Marshall and Rust, 1991; Rust and Marshall, 1996). However, it is difficult to fully understand the process of charge structure evolution and the origin of charge generation. At present, many cloud models coupled with charge separation mechanism have been explored to discuss the profiles of space electric field and charge structure in the evolution of a thundercloud (Takahashi, 1984; Rawlins, 1982; Helsdon *et al*., 2001; Mansell *et al*., 2005; Marshall *et al*., 2005). As all various electrification mechanisms are majorly dependent on environment temperature, hydrometeors concentration and size spectrum (Takahashi, 1978; Jayaratne *et al*., 1983; Saunders *et al*., 1991; Ziegler *et al*., 1991; Saunders and Peck, 1998), the validity of microphysics and hydrometeors is one of the key factors for simulating charge structure. Furthermore, the impact of aerosols on cloud microphysics and hydrometeors concentration and size spectrum are reasonably well understood (Khain *et al*., 1999; Yin *et al*., 2000). Thus, aerosols act as cloud condensation nuclei (CCN) perhaps have greatly effect on charge structure in thunderclouds.

An in-depth study of storm electrification requires numerical simulations. The parameterizations of charging mechanisms by which hydrometeors acquire charge are involved in cloud model. Most of related electrification parameterizations based on laboratory studies can be classified into inductive charging parameterization and non-inductive charging parameterization. The drop-ice interaction is considered as primary inductive mechanism (Aufdermauer and Johson, 1972; Moore, 1975), and the inductive charge transfer between two particles is connect with particles radius, the falling velocities, collision angle and environmental electric field (Mason, 1988). In addition, Non-inductive charge separation can be considered as a primary mechanism in thunderclouds, meanwhile several non-inductive parameterizations based on the laboratory results (Takahashi 1978; Gardiner *et al*. 1985; Jayaratne *et al*. 1983; Saunders *et al*., 1991; Brooks *et al*., 1997; Saunders and Peck 1998) are put forward to simulate charge separation via rebounding graupel-ice collisions. Although the comparison of these laboratory-based parameterizations in a full simulation model (with coupled dynamics and microphysics) has revealed significant differences between the results (Helsdon *et al*., 2001; Mansell *et al*., 2005), the sign and magnitude of electric charge separated during collisions between ice-phase particles highly generally depends on temperature, relative velocity of the collisions, hydrometeors concentration and the supercooled droplet size spectrum (Takahashi, 1978; Gardiner *et al*., 1985; Jayaratne *et al*., 1983; Saunders *et al*., 1991; Brooks *et al*., 1997; Saunders and Peck, 1998). In general, charge separation is closely related to microphysics and hydrometeors properties of thunderclouds. It is well-know that aerosols can change dynamical, microphysical, and hydrometeors properties of cloud (Khain *et al*., 1999; Yin *et al*., 2000). How aerosols affect electrification process in thunderclouds? However, at present very few previous simulation studies of aerosols effects have been performed in cumulus electrification model.

In recent years, considerable progress has been made in understanding aerosols, their microphysical properties, and the factors that enable them to act as cloud condensation nuclei (CCN) (Twomey, 1974; Albreht, 1989). As a result, aerosols exert a substantial influence on the microphysical properties of warm and cold clouds. Some observations and numerical simulations reveal that greater concentrations of aerosols result in the production of more small cloud droplets and reduced collision efficiencies, which act to delay the formation of raindrops (Brenguier *et al*., 2000; Durkee *et al*., 2000; Yin *et al*., 2000; Nakajima *et al*., 2001; Ramanathan *et al*., 2001; Feingold *et al*., 2003; Jirak and Cotton, 2006). On the other hand, aerosol concentrations have a substantial impact on mixed convective cloud (Khain *et al*., 1999; Lynn *et al*., 2005; Seifert and Beheng, 2006). Increase in the concentration of aerosol particles leads to higher vertical velocities; more super-cooled liquid water and increases in large ice-phase hydrometeor particles concentrations (Van den Heever *et al*., 2006; Yang *et al*., 2011). Aerosols, therefore, not only affect the microphysical development in clouds but also have influence on the physical characteristics of hydrometeor particles. As the mechanisms of thunderstorm electrification is intrinsically linked to microphysics and hydrometeor particles, the possible effects of aerosols particles on thunderstorm electrification should be studied with cloud models.

Some models have discussed below include aerosols and electrification process. A study by Takahashi (1984), who used a spectral bin dynamic model to study the effects of maritime (low) CCN and continental (high) CCN on electrification, suggested that aerosols might be responsible for significant enhancement for electrification for the continental CCN. Mitzeva *et al*. (2006) performed a 1D bulk-water model to investigate differences between the early electrical development of maritime and continental thunderstorms, and found that updraft enhancement, greater ice production, and stronger electrification with continental aerosol content compared to maritime. The influence of IN(ice nuclei) bacteria on thunderstorm structure and lightning formation has been studied using a regional atmospheric model, and just a relationship between lightning number and maximum cloud updraft was taken into account the storm dynamics (Goncalves, 2012). A recent study by Wang *et al*. (2011) revealed the impact of aerosols on precipitation and lightning under polluted aerosol and clean aerosol conditions with a two-moment bulk microphysical scheme. Although their results indicate electrification in thunderstorms is sensitive to aerosol concentration, for one thing they do not use detailed electrification scheme, which is the most important aspect of charge structure generation, for another the description of aerosol characteristic and activation process is incomplete.

From the above studies one can conclude that few attempts have yet been made investigate the aerosols effect on microphysics and electrification process in cloud models. The aim of this paper is to present sensitivity studies of aerosol concentration on thunderstorm microphysics and electrification. For this purpose, a two-dimensional cumulus model with detail cloud microphysics and electrification scheme is used. Numerical experiments are mainly tested for the relationship between aerosol concentration and distributions of space charge in thunderstorms.

Simulation method

This study used a 2-D Cartesian cumulus model, developed by the Chinese Academy of Meteorological Sciences (Hu and He, 1987). It is a non-hydrostatic cumulus model. Prognostic equations are included for momentum, pressure, potential temperature, and cloud droplet spectral width which is used to calculate the conversion of cloud droplet to rain. There are also conservation equations for mass ratio and concentration ratio of hydrometeors. The microphysics package is a multi-category, double moment scheme. It has five hydrometeor categories, which are cloud droplet, rain, ice crystal, graupel and hail. The hydrometeors are characterized as mass ratio (Q_x) and concentration ratio (N_x).

The main cloud physical processes are condensation and evaporation, collision, autoconversion, nucleation and multiplication, melting and freeze, and the model includes 27 kinds of microphysical processes of cumulus. The 27 kinds of microphysical processes are: condensation and evaporation of ice crystal, rain, cloud droplet, graupel, and hail; collision between cloud droplet and ice crystal, rain, graupel, as well as hail; collision between rain and ice crystal; collision between rain and graupel, and hail; collision between ice crystal and graupel, and hail; nucleation and multiplication of ice crystal; autoconversions of cloud-rain, ice-graupel, and graupel-hail; freeze of rain into graupel; melting of graupel, hail, and ice into rain; collection of ice, collection of rain, and welt growth of graupel.

To better understand the effect of aerosol on cloud microphysical processes, we made some improvements to the model. As a background field of the initial aerosol spectrum and concentration is added to this model, we fit a classic scheme for aerosol activation based on Köhler equation (Pruppacher and Klett, 1997), and the concentration of activation cloud droplets can replace the original constant (400 cm^{-3}). For simplicity, we assume the mass of activated cloud droplets is related to a minimum activation radius (Yin *et al*., 2000).

Under these conditions, we use a resolution of 250 m and time steps of 2 second to calculate the microphysical and electrification processes in 76 km ×20 km domain. It should be point out that in the present work we have ignored the process of lightning discharge, because the induced charge in lightning leaders results in a new and more complicated charge re-distribution (Tan *et al*., 2007; Tao *et al*., 2009), which probably go against the investigation on the influence of aerosol concentration on the charge in thunderclouds. In addition, some important methods are discussed as follows.

The electrification scheme

An electrification scheme must be included in numerical cloud model to generate charge acquired by hydrometeors. Similar to Mansell *et al*. (2002), the electrification parameterizations include inductive charge separation between grapuel/hail and cloud droplet under the external electric field and non-inductive mechanisms between grapuel and ice crystal under the coexist of ice particles and supercooled water.

Aerosol spectral distribution

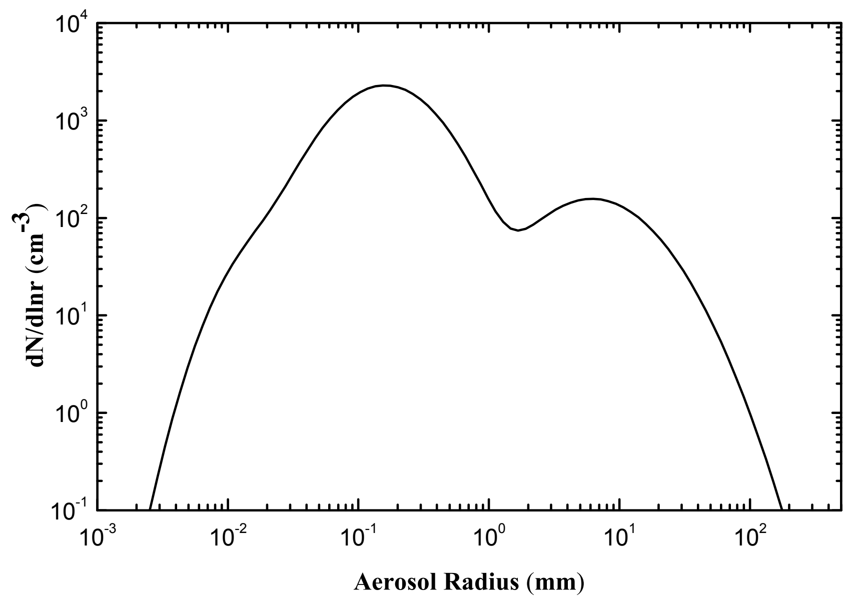


Fig.1. initial number distribution functions of aerosol particles used in the simulations

The aerosol distribution of Hobbs *et al*. (1985) was fitted by superimposing three log-normal distribution function. In Equation (1), the subscript i=1, 2, or 3 represents the three modes. r_N is the radius of aerosol particle, and n_i denotes the aerosol number concentration in mode i, the ratio of n_i for three log-normal distribution was 72: 4430: 450. σ_i (σ_1 =1.8, σ_2 =2.16, σ_3 =2.40) is the geometric standard deviation representing the width of the particle size, and the three distributions had respective geometric mean radius (R_i) of 0.02, 0.16, 6.15μm. The parameters of the distributions were based on Fig.1 of Leporini *et al*. (2004).

$$\frac{dN}{d\ln(r_N)} = \sum_{i=1}^{3} \frac{n_i \exp\left(-\frac{\ln^2(r_N / R_i)}{2\ln^2(\sigma_i)}\right)}{\sqrt{2\pi}\ln(\sigma_i)} \tag{1}$$

Activation of aerosols

The aerosol particles begin to grow by absorption of water vapour. The activation of aerosol particles to form cloud droplets depends on the assumed aerosol composition and local supersaturation. In this study, the composition of aerosol particles is assumed to be isotropic homogeneous distribution of sulfate particles. Aerosol particles of a certain size are activated when the radius of growing aerosol particles at each grid point exceeds the critical activation radius (r_{min}) determined by the Köhler equation (Pruppacher and Klett, 1997):

$$\Delta S = \frac{A}{r} - \frac{Br_d^3}{r^3} \tag{2}$$

Where, r_d is dry aerosol particle radius and r is wet aerosol particle radius, A is the coefficient of the curvature effect and B is the coefficient of solute effect.

$$A = \frac{2\delta}{\rho_w R_v T} \tag{3}$$

$$B = \frac{i\varphi_s \varepsilon_m \rho_N M_w}{\rho_w M_N} \tag{4}$$

$$r_{\min} = \left(\frac{4A^3}{27B}\right)^{1/3} \cdot \Delta S^{-2/3} \tag{5}$$

Where, δ is the surface tension of the solution drop, ε_m is the fraction of water-soluble material of an aerosol particle, i is the number of soluble particles molecules (for $(NH4)_2SO_4$, i=3). M_w and M_N are the molecular weights of water and aerosols. ρ_w and ρ_N are the densities of water and aerosols, respectively.

After reaching the critical sizes, the number concentration of cloud droplets is associated with activated aerosol concentration. In addition, based on Yin et al. (2000), we assume a correspondence relation between cloud droplets quality and the certain size. *Mc* is cloud droplets quality and is given by:

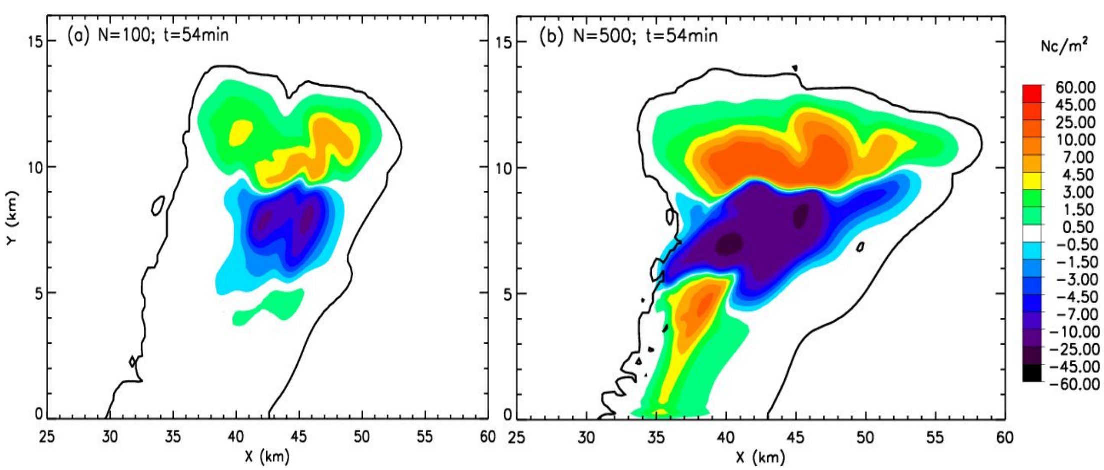
$$M_c = \rho_w \cdot \frac{4}{3}\pi (kr_{\min})^3 \tag{6}$$

In which, a factor k is used to calculate the ratio of initial sizes of the droplets to aerosol particle radius based on Kogan (1991).

Simulation results

Effects of aerosol concentration on electrification

As previously discussed in last section, four cases are performed for charge structure in thunderstorm cloud. The focus here is on the dominant aerosol concentrations relevant to charge characteristics. The mature stage of storm at 54 min depicts a triple charge structure with a lower positive charge below a normal dipole, and comparing these figures in Fig.2 reveals that when the aerosol concentration increases, the charge structure of four cases consistently keep as a triple, but the charge density distribution shows significant differences. The estimated charge magnitudes of upper positive charge region (primarily above 9 km level), middle negative charge region (between 5 km level and 9 km level) and lower positive charge region (below 5 km level) are defined as Q_{up}, Q_{mn} and Q_{lp}, respectively. Assuming that the charge density structure of each plane across Z-axis is same as that of X-Z plane in thunderstorm, the horizontal radius of thunderstorm of four cases is 1 km. As shown in Table 1, the absolute charge magnitude in middle negative region is larger than values of upper or lower positive charge region. Furthermore, Q_{mn} and Q_{lp} increase monotonically as aerosol concentration rises from 100 to 3000 cm^{-3}, but Q_{up} shows different characteristic that positive charge on upper region begins to decrease when N increases from 1000 to 3000 cm^{-3}.



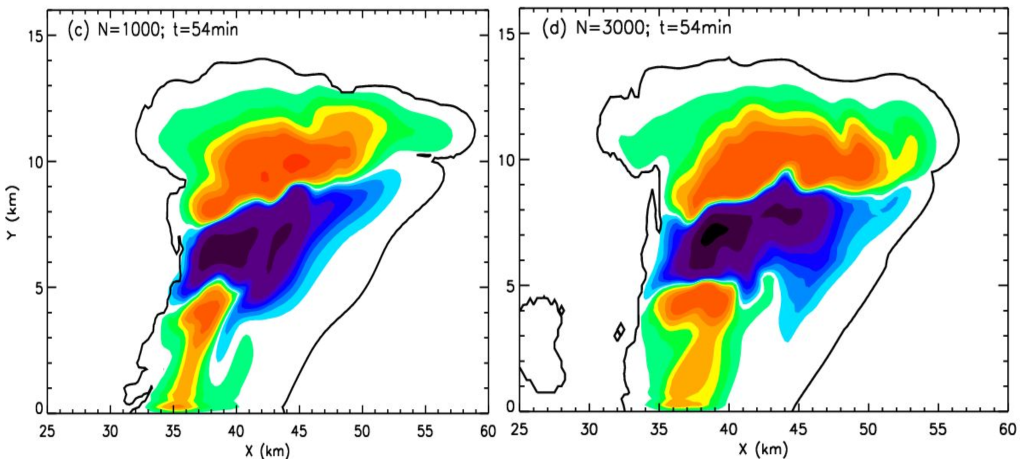


Fig.2. Charge structure in the mature stage of thunderstorm for initial aerosol concentrations: (a) 100 cm^{-3}, (b) 500 cm^{-3}, (c) 1000 cm^{-3}, (d)3000 cm^{-3}.

Table 1.Charge values of upper positive charge region(Q_{up}), middle negative region (Q_{mn})and lower positive charge region(Q_{lp})

Case	Q_{up}(C)	Q_{mn}(C)	Q_{lp}(C)
N=100	88	-100	5
N=500	275	-371	73
N=1000	365	-471	76
N=3000	359	-489	117

In this study, NI (non-inductive) and inductive charging rates contribute to the total charge structure, which is consistent with a model study by Mansell (2005). Graupel collides with ice crystals, charging the ice crystals positively and graupel negatively via NI charging mechanism. The lighter ice crystals that gain positive charge are carried upward into the cloud anvil by updraft, and they mostly reside between 9 km level and 13 km level (Fig.8a), then from the upper positive charge region. It is obvious from Fig.8a that around 10 km level space charge from ice crystals reaches a peak. These peaks are 3.5 nC m^{-2} (100), 4.9 nC m^{-2} (500), 9.9 nC m^{-2} (1000), and 7.1 nC m^{-2} (3000), respectively. The difference in the content, concentration and diameter of ice crystals and graupel among four cases can be found in Table 2. The magnitude of charge produced from NI mechanism is mainly associated with diameter of ice crystals and mass concentration of ice crystals and graupel, and therefore smaller of ice crystals will weaken NI charging rate when aerosol concentration enhance. However, this impact is much weaker than ice-phase particles concentration influence because of little change in diameter of ice crystals (See Table 2). In comparison, the influence of ice-phase hydrometer concentration is much stronger. The mass concentration of ice crystals and graupel make the greatest contribution to charge generation. It is obvious from Table 2 that the mean mass concentration of ice crystals reached their maximum when the initial aerosol concentration is 1000 cm^{-3} and mean mass concentration is 0.20 g kg^{-1} (Relatively high). This may be one reason that charge in upper region in 1000 cm^{-3} case is stronger than other three cases.

Table 2.The main results obtained from four experiments (at 54 min)

Hydrometer parameters	N=100	N=500	N=1000	N=3000
Mean. mass of cloud droplet (g/kg)	0.12	0.16	0.24	0.31
Mean. number of cloud droplet (cm^{-3})	11.0	30.0	103.2	846.3
Mean. diameter of cloud droplet (μm)	56.2	35.8	18.5	11.2
Mean. mass of graupel (g/kg)	0.15	0.19	0.20	0.21
Mean. number of graupel (I^{-1})	9.2	15.1	17.8	18.5
Mean. diameter of graupel (mm)	3.8	3.2	3.0	3.0
Mean. mass of ice crystal (g/kg)	0.32	0.36	0.38	0.30
Mean. number of ice crystal (I^{-1})	28.5	42.3	49.7	30.4
Mean. diameter of ice crystal (μm)	98.4	96.5	95.2	95.4

The main middle level negative charge region is mainly attributed to charging of graupel and cloud droplets (Fig.2b and c). NI graupel charging affected by gravitationally separating maintains negative charge at about 7-10 km level. In addition, with the polarity reversal of vertical electric field at about altitude of 7 km (Fig.8d), graupel is negatively charged at about 6-10 km level and positively charged at about 3-6 km level via inductive charging mechanism, which also leads to cloud droplets positively charged at about 7-12 km level and negative charged between 4 km level and 7 km level. Therefore, the merger in different polarity charge acquired by graupel and cloud droplets is primarily responsible for sedimentation of charge on middle region. The enhancement of hydrometeors production and vertical electric field arising from increasing aerosol concentration can contribute to greater inductive charge between grapuel and cloud droplets, which can be seen in Fig.2b,c and d. In addition, the reduction of grapuel and cloud droplet diameter at very high aerosol concentration is a matter of reduced inductive charge production, but this impact is insignificant.

Inductive graupel charging is primarily responsible for the lowest positive charge region (Fig.2b) where raindrops is falling (not shown), and raindrops below the melting level acquire weaken positive charge via phase transformation of hydrometeor particles, which is consist with observation by takahashi (2010). The enhancement of graupel concentration due to increasing aerosol concentration is likely responsible for the stronger positive charging on the lowest region.

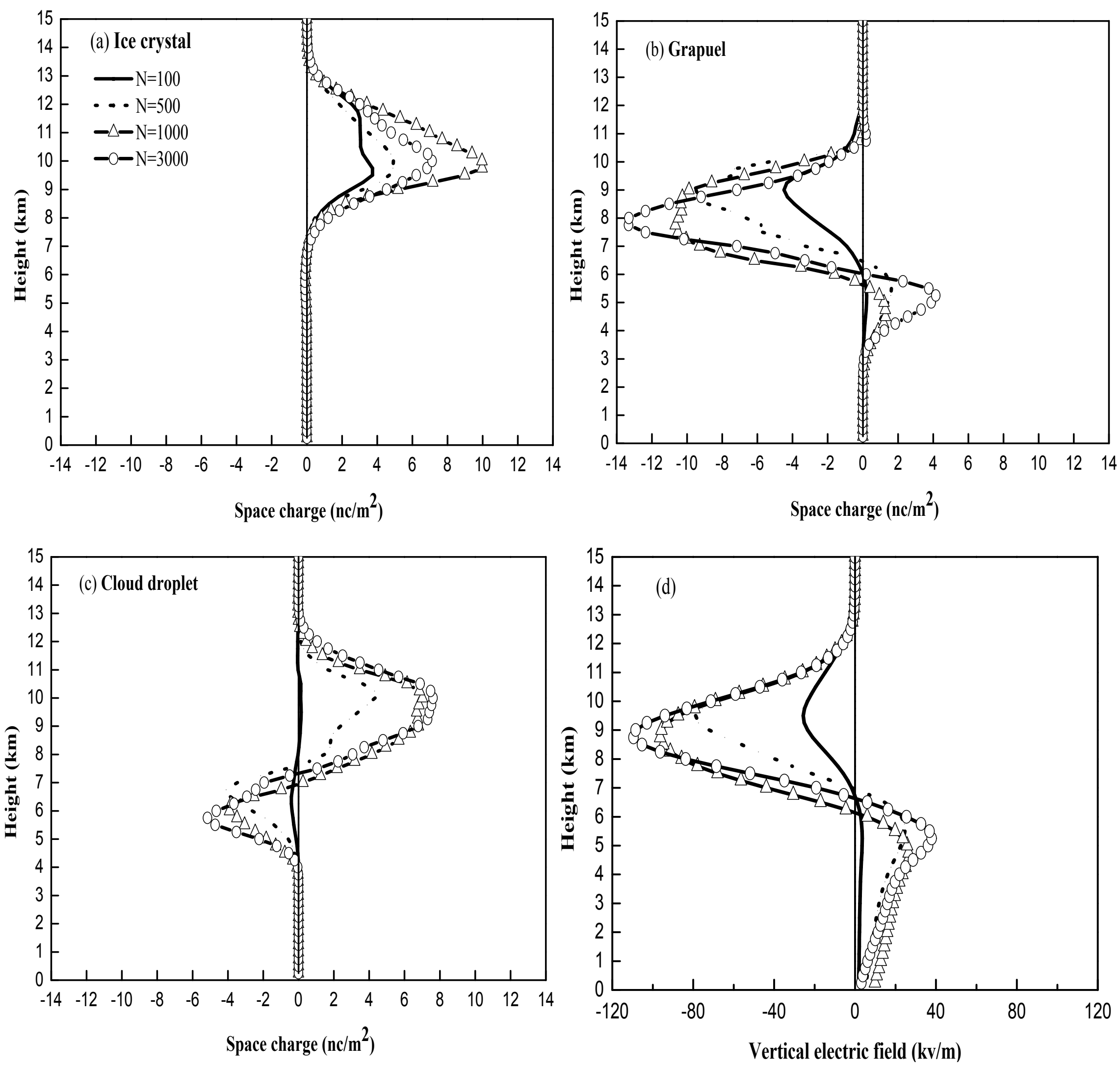


Fig.2. Space charge from ice crystals (a), grapuel (b) and cloud droplet (c). The vertical electric field inside thunderclouds in four cases (d). Further, an upward-directed electric field is defined as positive.

Electrification with high aerosol concentration

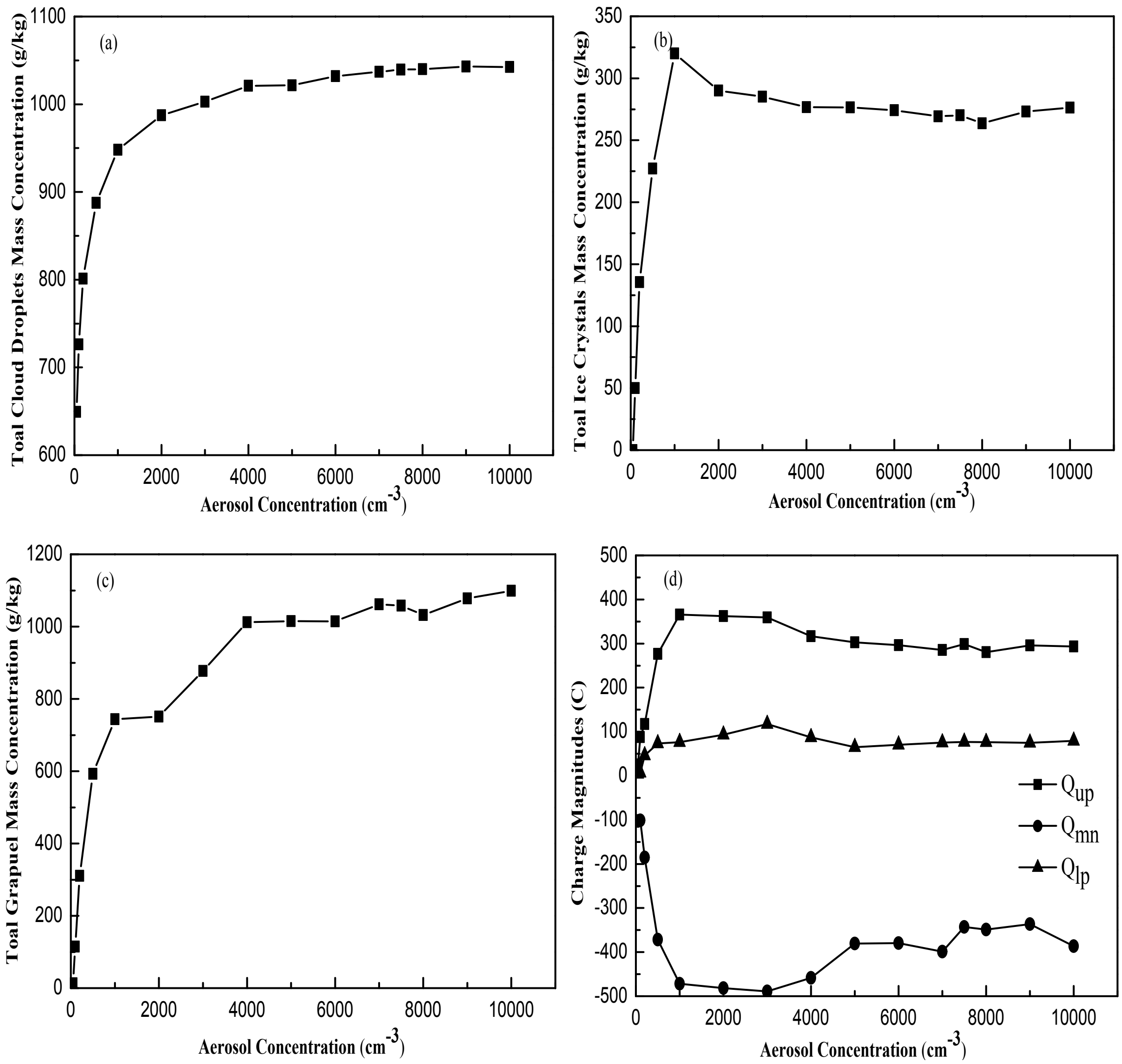


Fig.3.Total cloud droplets (a), ice crystals (b), graupel (c) mass concentration and the estimated charge magnitudes in upper positive charge region, middle negative charge region and lower positive charge region for varying aerosol concentration(d).

From the analysis in the previous section, we know that the production of ice crystals at higher altitudes is reduced by vapor competition at aerosol concentration of 3000 cm^{-3}, which is closely associated with the decrease of charge acquired by ice crystals. Of interest here is on the dominant charge characteristics relevant to higher concentration aerosols (above 3000 cm^{-3}). The total cloud droplets (Fig.3a) and grapuel (Fig.3c) mass concentration follows the trend of rapidly increasing at lower aerosols (50-4000 cm^{-3}), with slowly increasing at higher aerosols (4000-10000 cm^{-3}). Therefore, when aerosol concentration exceeds 3000 cm^{-3}, the increase in cloud droplets concentration seems insensitive to aerosol concentration. This feature seems to be due to all simulation cases under the same relative humidity condition, and water vapor content in cloud is certainly, which restricts more small cloud droplets further condensation growth. Since collision between graupel and cloud droplet is always a greater source to graupel mass concentration than the initial source of auto-converted ice crystal and cloud droplet (not shown), the production of graupel is mainly depends on graupel-cloud droplet collisions, the variation of graupel concentration is correlated with cloud droplets concentration. On the other hand, In Fig.3b, vapor competition can be explained the reduction in ice crystal at aerosol concentration mediated from 1000 cm^{-3} to 3000 cm^{-3}, but when aerosol concentration further increases, the total ice crystals mass concentration at 34 min (time for peak value) will be a stable magnitude of about 275 g kg^{-1}, which mainly results from the similar supply of water vapor for ice crystals production.

The charge structure in mature stage keeps as a normal tripole for all simulation cases. From Fig.3d we can conclude that the total charge separation increases from 50 cm^{-3} to 3000 cm^{-3}, although a small decline in upper positive charge region at aerosol concentration of 1000-3000 cm^{-3}would be caused by ice crystal concentration decrease. The increasing charge separation produces stronger electric field (see Fig.3d), which is more likely to trigger the occurrence of lightning. This simulation is consistent with observational evidences that aerosol may enhance lightning production (Yuan *et al*. 2011 and Wang *et al*. 2011). Above aerosol concentration of 3000 cm^{-3}, the estimated charge magnitudes in upper positive charge region, middle negative charge region and lower positive charge region with value of 280 C, -385 C and 80 C, respectively are slightly less then that at aerosol concentration of 3000 cm^{-3}, and the estimated charge magnitudes have no obvious change. The decrease ice crystals is likely to hold the answer to non-inductive charge separation reduce, which is contributed to the weaker initial environmental electric field (not shown). Aside from electric field, the factor that graupel and cloud droplets concentration increase insignificantly at very high aerosol concentration would affect electrification (see Fig3a and c). Under these condition, the inductive charge separation depends on graupel-cloud droplet collisions would be reduced.

In generally, the charge separation increases as aerosol concentration mediated from 50 cm^{-3} to 3000 cm^{-3}, so the peaks in charge of thunderstorm cloud is at the aerosol concentration of about 3000 cm^{-3}. When aerosol concentration exceeds 3000 cm^{-3}, the charge separation which weakens to a certain magnitude is insensitive to aerosol concentration.

Conclusions

The simulations have been performed to investigate the impact of varying initial aerosol concentration on the thunderstorm charging. The analyses of results demonstrate that aerosol concentrations have a significant influence on the thunderstorm cloud microphysical processes and electrification. From these results one can conclude the following:

Charge structure of thunderclouds in mature stage always keeps as a normal triople when aerosol concentration increases. Charge separation tends to increase as aerosol concentration rises from 50 cm^{-3} to 3000 cm^{-3}. The enhancement of inductive charge from collisions between graupel and cloud droplets, arising from graupel and cloud droplets concentration and vertical electric field increase, is a significant factor in enhance of middle negative charge and lower positive charge in thunderclouds. However, the reduction of ice crystals concentration in the range of 1000-3000 cm^{-3} slightly inhibits charge production via non-inductive mechanism. Under this condition, the magnitude of upper positive charge mainly attributed to non-inductive charging of ice crystals have a slightly reduce. A little change in hydrometeor concentration at very high aerosol concentration (above 3000 cm^{-3}) can be attributed to the stability of space charge magnitude. In general, variation of charge in thunderclouds implying that the peak of charge magnitude in thundercloud is at the aerosol concentration of about 3000 cm^{-3}.

This present study reveals that the cloud microphysical and electrification properties depend on the aerosols concentration under the same initial dynamic and thermodynamic conditions, but the effect of aerosols concentration on charge separation in thundercloud is non-linear. Although the association between lightning activity and aerosol has received further strong support from studies (Yuan et al., 2011) which demonstrate strong and quantifiable relationships – obtained from the analysis of data from observations, the key electrification processes associated with further high aerosol concentration are still short of anecdotal evidence. Furthermore, it is believed that aerosol can act as ice nuclei (IN). Because cloud-ice nuclei (IN) interaction is increasingly recognized as one of the factors influencing the microphysical structure of clouds (Levin, 2005; Teller, 2006; Khain and Blyann, 2009), and ice crystal (nucleation of IN) is of crucial importance in thunderstorm electrification, this process may have an important impact on electrification properties in thunderclouds. Further aspects of this problem will be addressed in forthcoming studies.

Acknowledgments

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