

New particle formation by ion-induced nucleation during dissipation stage of thunderstorm

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A case of new particle formation observed during dissipation stage of a thunderstorm at a tropical station, Pune, India on 3 June 2008 is reported. The flash rate and rainfall intensity increased as high as 110 flashes per 5 minutes and 150 mm hour⁻¹ respectively during the active stage of thunderstorm, and then gradually decreased during the dissipation stage. The number concentration of particles in the size range of 10–100 nm sharply increased from 350 particles cm⁻³ to ~8000 particles cm⁻³ during the dissipation stage of a thunderstorm and grew to larger diameter subsequently. Observations suggest that the atmospheric conditions such as (i) reduced background aerosol concentration after heavy rain, (ii) high humidity condition, and (iii) increased ion concentration during the dissipation stage by corona discharges, favoured generation of new particles by ion-induced nucleation (IIN). Observations also suggest that generation of unipolar ions by corona discharges may be more favourable for IIN and subsequent growth of the particles.

1. Introduction

Events of aerosol nucleation have been observed at various parts of the world at different atmospheric conditions. The mechanism and atmospheric conditions controlling the onset of such events are not yet clearly understood. The gas-to-particle conversion (GPC) process, in which small clusters of molecules form through binary homogeneous nucleation of sulphuric acid and water and subsequently grow to particles larger than 3 nm, could not alone explain the nucleation rates observed at various parts of the world (Kulmala *et al* 2004). Wilson (1895) had shown for the first time that increased ionization could trigger the process of new particle formation. Cluster ions can grow to form new particles and rate of such new particle formation will be proportional to ion concentration or

the ionization rate in the atmosphere (Dickinson 1975). Later, many studies have identified ion-induced nucleation (IIN) as one of the most important source of atmospheric particles at some locations. Laakso *et al* (2002) have suggested that new particle formation by IIN process can occur under some favourable atmospheric conditions, like low temperature, high relative humidity, high ion production rate, and low concentrations of pre-existing particles. The findings of Lee *et al* (2003) indicate that, at upper troposphere and lower stratosphere, because of favourable conditions such as relatively high ion-production rates by galactic cosmic rays, low temperatures, and relatively low surface areas of pre-existing aerosols, ion-induced nucleation process is a dominant process for new particle formation. Yu (2006) has demonstrated using a kinetically consistent ion-mediated

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nucleation model that ions can lead to significant particle formation not only in the upper troposphere but also in the lower troposphere including boundary layer.

Here we present an event of new particle formation observed in the dissipation stage of a thunderstorm. Role of increased ionization during dissipation stage of thunderstorm due to point discharges have also been discussed.

2. Observatory, instrumentation and data

The Atmospheric Electricity Observatory is situated at Pune (18°32'N, 73°51'E, 573 m AMSL), India, on the lee side of the Western Ghats about 100 km inland from the west coast of India. Measurements of atmospheric electric field, Maxwell current density, aerosol size-distribution, rain intensity and rate, along with meteorological parameters such as temperature, humidity, wind speed and direction are being made at the surface level in this observatory.

Atmospheric electric field (E) is measured by a vertical field mill as described by Kamra and Pawar (2007). The time constant of the field mill is adjusted to 0.1 s. The field mill can measure the electric field in the range ± 15 kV/m and the response of the field mill in this range is found to be linear during the calibration. The sensitivity of the field mill is ± 2 V/m and the noise level is well below ± 2 V/m. Aerosol number concentration and size-distributions in the mid-point diameter range of 3–113 nm have been measured using a Scanning Mobility Particle Sizer (SMPS, Model 3936, TSI Inc., USA). The SMPS sequentially scans for each channel starting from 3 to 113 nm using a differential mobility analyser (DMA). Even though SMPS has the ability to measure particles down to 3 nm size, the transfer function (the probability that a particle of electrical mobility Zp entering the DMA, will exit the DMA in a given monodisperse aerosol flow without diffusion loss) used in this measurement derived by Knutson and Whitby (1975) and this equation do not account for diffusion losses. This is very critical in smaller particles size even in the fully developed laminar flow. It has been shown that diffusion losses and broadening of transfer function can create large uncertainty, especially, in the lower size bins of measurements (Kousaka *et al* 1985; Mamakos *et al* 2007). Therefore, we have used particles above 10 nm size in this study. Time resolved size-spectra of rain drops from 0.3–20 mm diameter sizes is measured by a disdrometer (OTT Parsivel, Germany) and the rain rate and intensity are calculated from this spectrum.

3. Results

A thunderstorm developed in the vicinity of our observatory at about 1600 hrs local time (LT) on June 3, 2008 and lasted for about 2 hours. Figure 1 shows the meteorological parameters observed on that day during the thunderstorm period. The winds which were initially about 5 m sec^{-1} increased to $10\text{--}15 \text{ m sec}^{-1}$ during active stage and again decreased to less than 3 m sec^{-1} at about 1800 hrs LT in the dissipation stage. Air temperature sharply decreased by about 10°C during the thunderstorm period and relative humidity was more than 90%. As shown in the figure 2(a), with the onset of thunderstorm, electric field increased to 2 kVm^{-1} and lightning activity started at about 1620 hrs. The flash rate and rain intensity are shown in figure 2(b). Lightning flash rate increased very rapidly, reached to 120 flashes per 5 minutes during active stage of thunderstorm, and then decreased gradually to less than one flash per 5 minutes at about 1800 hrs. Precipitation started at about 1700 hrs LT and the rain intensity sharply increased to 150 mm hr^{-1} in the active stage of thunderstorm and then decreased gradually to 3 mm hr^{-1} in the dissipation stage, at about 1800 hrs LT (figure 2b). The total aerosol concentration along with 5-minute average of electric field is shown in figure 2(c). The concentration of aerosols decreased from about 2000 particles cm^{-3} just before the start of thunderstorm to less

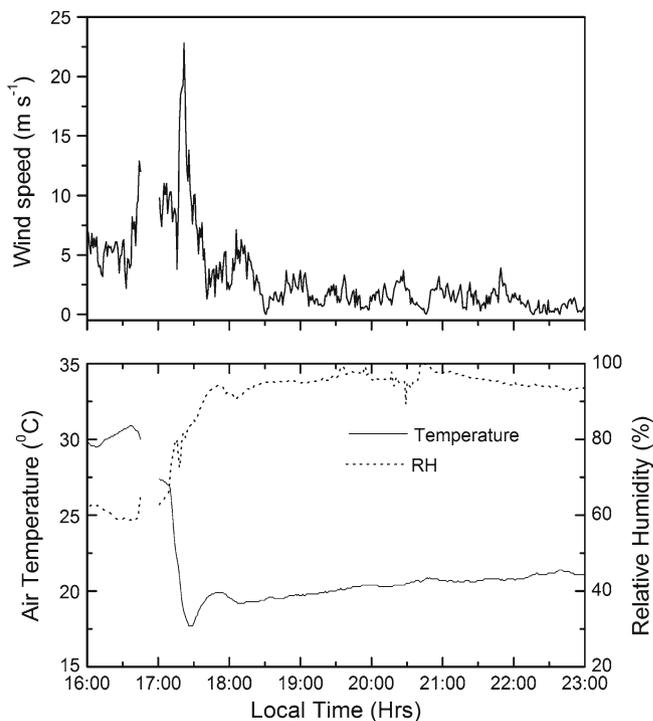


Figure 1. Plots of temperature, humidity and wind speed during the thunderstorm on June 3, 2008.

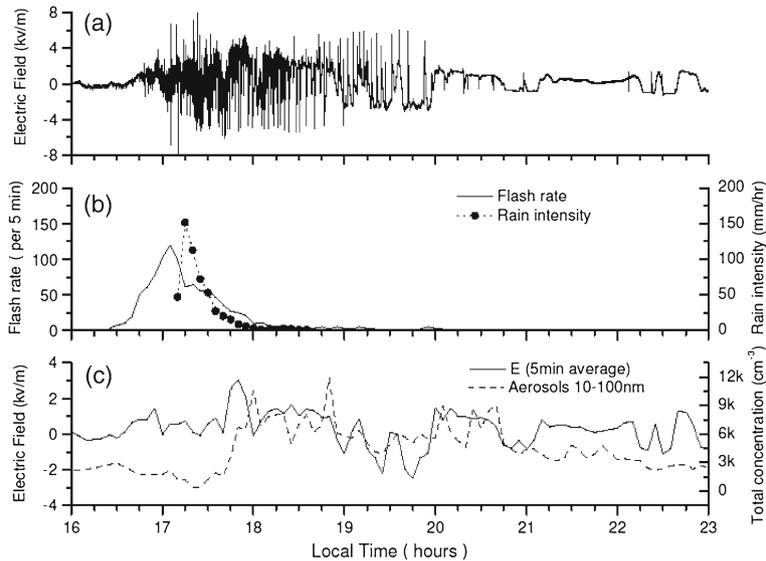


Figure 2. Variations of (a) electric field, (b) lightning flash rate and rainfall intensity, and (c) 5-minute averaged electric field along with total concentration of aerosols in the size range 10–100 nm during the period of thunderstorm.

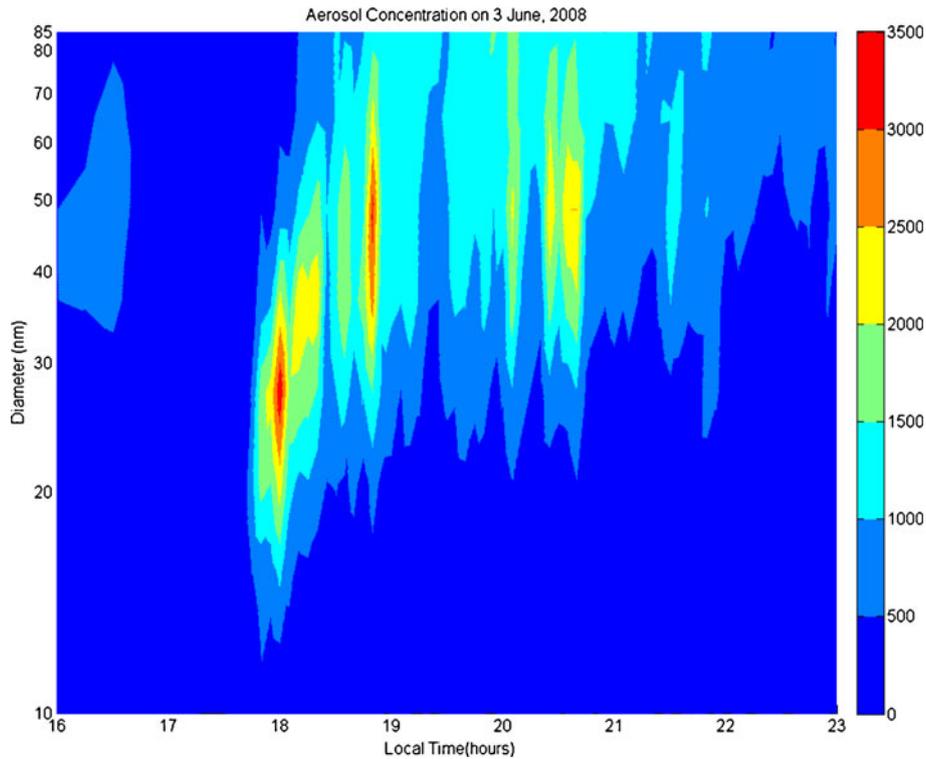


Figure 3. Temporal variation of aerosol number-size distribution during the thunderstorm period.

than 500 particles cm^{-3} during heavy precipitation. Sudden increase in aerosol concentration was observed with the cessation of rainfall, at about 1800 hrs (figure 2c). Figure 3 shows the time evolution of size distributions of aerosols during this thunderstorm. Removal of all sizes of aerosols by precipitation during active stage of thunderstorm can be seen clearly in this figure. Sudden burst in ultrafine mode particles at 1800 hrs LT and

subsequent growth of these particles also can be seen. It should be mentioned here that less particle concentration in the size range 10–20 nm, might be due to diffusion losses as explained by Mamakos *et al* (2007). The average growth rate from 1800 to 1900 hrs LT was about 20 nm/hour, which decreased sharply after 1900 hrs LT. As can be seen in figure 3, the growth rate in the first half an hour was as high as about 35 nm/hour and such

a high growth rate has been reported very rarely. Kulmala *et al* (2004) have reviewed the cases of new particle formation all over the world. As they have shown, the growth rates lie between 1 and 20 nm/hr in most of the places. However, O'Dowd *et al* (2002), over the western coast of Ireland, have reported growth rate as high as 200 nm/hr.

Another case of new particle formation was observed during a thunderstorm on June 4, 2008. On June 4, 2008, a thunderstorm was developed near to our observatory at about 1530 hrs LT. This thunderstorm was not as severe as on June 3, 2008. Lightning frequency was less initially at 1550 hrs LT, became intense and increased up to 27 lightning flashes per 5 minutes at about 1700 hrs LT and decreased gradually in dissipation stage at about 1720 hrs LT. Precipitation started at about 1650 hrs LT and rain intensity increased to 15 mm hr⁻¹ in the active stage of thunderstorm and then decreased gradually in the dissipation stage at about 1720 hrs LT. Figure 4 shows the time evolution of size distribution of aerosols during this thunderstorm. As shown in this figure, there was sudden increase of ultrafine mode particles at about 1720 hrs LT just after dissipation stage. It can be seen from figure 4, the subsequent growth of the ultrafine particles into higher diameter; however, the growth rate is not as high as observed in case of thunderstorm observed on June 3, 2008 (figure 3).

4. Discussion

The electric field at the tip of sharp, grounded and elevated objects below thunderstorms can be as high as many hundred times than that of the ambient electric field. If the magnitude of the ambient electric field exceeds a few kV m⁻¹, it produces electrical discharges at the tip of elevated objects known as coronae discharges. Livingston and Krider (1978) and Krider and Musser (1982) have shown that due to frequent lightning, time averaged electric field and hence corona current could be very small during active phase of thunderstorm. However, in the dissipation stage, time averaged electric field value becomes high because of reduced lightning frequency, which enhances the corona current. Pawar and Kamra (2009) have shown that the space charge released into the atmosphere by corona currents from the ground can increase the air-conductivity of the atmosphere near to earth surface by more than an order of magnitude during the dissipation stage of thunderstorm.

Our observations clearly show an event of new particle formation and subsequent growth of those particles (figure 3). Since ion production rate

determines the amount of ions available for nucleation, it has been considered as the main factor for initiation of ion-induced nucleation. Even though measurement of ions was not made during this thunderstorm, earlier observations of Pawar and Kamra (2009) at this place and at many other places (e.g., Standler and Winn 1979; Standler 1980; Livingston and Krider 1978; Krider and Musser 1982) clearly suggest increase in ionization rate by almost an order, near to earth surface during dissipation stage. It should be noted here that maximum value of averaged electric field has reached more than +3 kV/m just before the start of this event. Laakso *et al* (2002) have shown that some favourable conditions, like low temperature, high relative humidity (RH), high ion production rate, and low concentrations of pre-existing particles are essential to initiate ion-induced nucleation process. As shown in figure 2(c), the background aerosol concentration has reduced to a very low value due to heavy rainfall just before the start of this nucleation event. In addition, as shown in figure 1, higher humidity of about 90% and low temperature prevailed during this event. Under such favourable atmospheric conditions, the observed new particle formation can be attributed to ion-nucleation process. New particle formation events cannot be observed unless the source rate of condensable vapours is sufficiently high (Kulmala *et al* 2004). Murugavel and Chate (2009) have estimated the concentration of vapour and its source rate at this location and have found that the values are high enough to trigger the nucleation bursts.

In our observation, electric field was of positive polarity at the time when new particle event was observed. Therefore, ions released in the atmosphere due to corona discharges should be of positive polarity. Some studies suggest that the negative ions, rather than positive ions contribute significantly to ion-induced nucleation process. However, measurements made by Hirsikko *et al* (2007), Arnold (2008) and laboratory experimental study by Rabeony and Mirabel (1987) suggest that positive and negative ions contribute almost equally to new particle formation. It should be noted here that during the processes of corona discharge, unipolar ions are released in the atmosphere. Arnold (2008) has shown that ion concentration depends on ion production rate and the ion-ion recombination coefficient ($N = (Q/\alpha)^{1/2}$), where Q is ionization rate and α is ion-ion recombination coefficient). Enghoff and Svensmark (2008) have also shown that ion nucleation process is not limited by the ion production rate, however the limiting factor is the rate of cluster production relative to the loss rate of ions by recombination and scavenging by larger clusters. In the case of unipolar ion generation, recombination coefficient

α would be negligible compared to bipolar ionization because of very low concentration of other polarity ions. In addition, the lifetime of cluster ions can be higher in such unipolar ion environment than in normal environment. The increase in life time of cluster ions can increase the growth rate of new particles. Higher growth rate of 35 nm per hour observed by us in first half an hour of new particle formation event is significantly higher than the values reported earlier by Kulmala and Kerminen (2008).

Kulmala *et al* (2004) have reported that the growth rate lies in the range of 1–10 nm h⁻¹. Some cases of growth rates as small as 0.1–1.0 nm h⁻¹ have been observed in remote locations (e.g., Vehkamäki *et al* 2004), whereas growth rate larger than 10 nm h⁻¹ has been reported in polluted environments (Kulmala *et al* 2005; Laaksonen *et al* 1995; Stolzenburg *et al* 2005; Hamed *et al* 2007; Wu *et al* 2007). Very high growth rates in excess of 100 nm h⁻¹ can be reached in plumes and in coastal zones intermittently (e.g. O'Dowd *et al* 2007). Our observation of growth rate of 35 nm h⁻¹ cannot be explained by conventional nucleation process and needs some other mechanism as proposed by us.

Another case of observation of new particle formation occurred on June 4, 2008 is not as systematic as observed on June 3, 2008, as shown in figure 4. However, we do observe abrupt increase in ultrafine particles and their subsequent growth into bigger diameters. This increase in ultrafine particles was observed in the dissipation stage of thunderstorm when lightning frequency reduced and electric field was high. These observations (figure 4) strengthen our hypothesis that low background aerosol concentration due to washout by rain and increased ion concentration due to corona discharges during dissipation stage of thunderstorm can trigger new particle formation via ion nucleation process.

Increase in concentration of particles less than 50 nm during dissipation stage is a common occurrence in thunderstorms observed in Pune. One such case is plotted in figure 5 where time evolution of electric field and aerosol concentration particles below 50 nm observed during a thunderstorm on May 22, 2008. As shown in figure 5, the particle concentration, which was about 3000 particles per cm³ before thunderstorm, reduced to 1500 particles per cm³ after the start of precipitation and increased sharply during the dissipation stage of thunderstorm. Such observations clearly suggest that the process of new particle formation occurs below thundercloud when conditions such as low background aerosols and high ion concentrations are fulfilled. The newly formed particles can increase the aerosol concentration in the surrounding area.

It should be noted here that both the cases of new particle formation presented here occurred during evening time when sun radiation is less. Ayers *et al* (1980) have pointed out that because of its low vapour pressure, generally nucleation is initiated by sulfuric acid. Further as H₂SO₄ forms from SO₂ and OH, and the majority of OH sources are the photochemical production of ozone and water under ultraviolet (UV) radiation, night-time nucleation from this process is not possible. However, in contrast to the current prevailing assumption that aerosol nucleation takes place only during the daytime and typically from sulfuric acid, many observations indicate efficient formation of new particles in night-time. For example, Hermann *et al* (2003) observed frequent ultrafine particles at night in a large fraction of aerosol samples in the subtropical and midlatitude tropopause regions. Mauldin *et al* (2003) observed H₂SO₄ and ultrafine particles at night over the Pacific Ocean. Observations made in and near orographic clouds by Wiedensohler *et al* (1997) and Meters *et al* (2005) also showed night-time nucleation events. Observations by Lee *et al* (2008) indicate new particle formation during the night-time in the troposphere under low condensation sinks. The continuous ground-based measurements in Tumberumba, Australia also indicate night-time new particle formation in the clean environment (Lee *et al* 2008). Rissler *et al* (2006) and Russell *et al* (2007) present instances of new particle formation during night-time takes place in a wide range of geographical locations. Lee *et al* (2008) have suggested that convection can bring higher concentrations of aerosol precursors and insoluble organic compounds from lower altitudes and contribute to new particle formation in the upper troposphere. Brown *et al* (2006) have suggested that SO₂ conversion by night-time oxidants (e.g., NO₃) to H₂SO₄ is also possible, in polluted plumes where sulfur and nitrogen oxides are abundant. O'Dowd *et al* (2002) and Bonn and Moortgat (2003) have hinted that the low volatility condensable organic compounds might possibly generate new particles even in the absence of H₂SO₄. These organic compounds can form via ozone or NO₃ in the absence of UV and OH during the night-time. Our observations also suggest that low background aerosol concentration and high ion concentration can trigger new particle formation process via ion nucleation.

We propose that during dissipation stage of thunderstorm when ion concentration increases more than an order due to corona discharges, the new particle formation can occur by ion nucleation process. Williams (2009) and Williams and Heckman (1993) have suggested that corona current can be a dominant contributor for global electric circuit other than lightning. Williams and

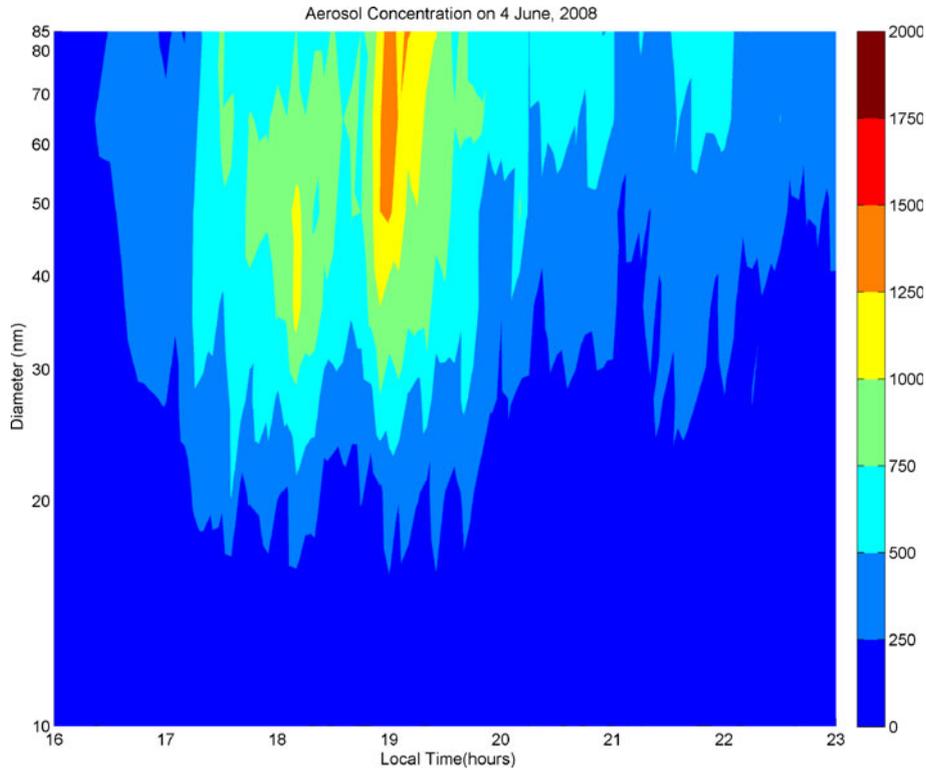


Figure 4. Same as in Figure 3 but on June 4, 2008.

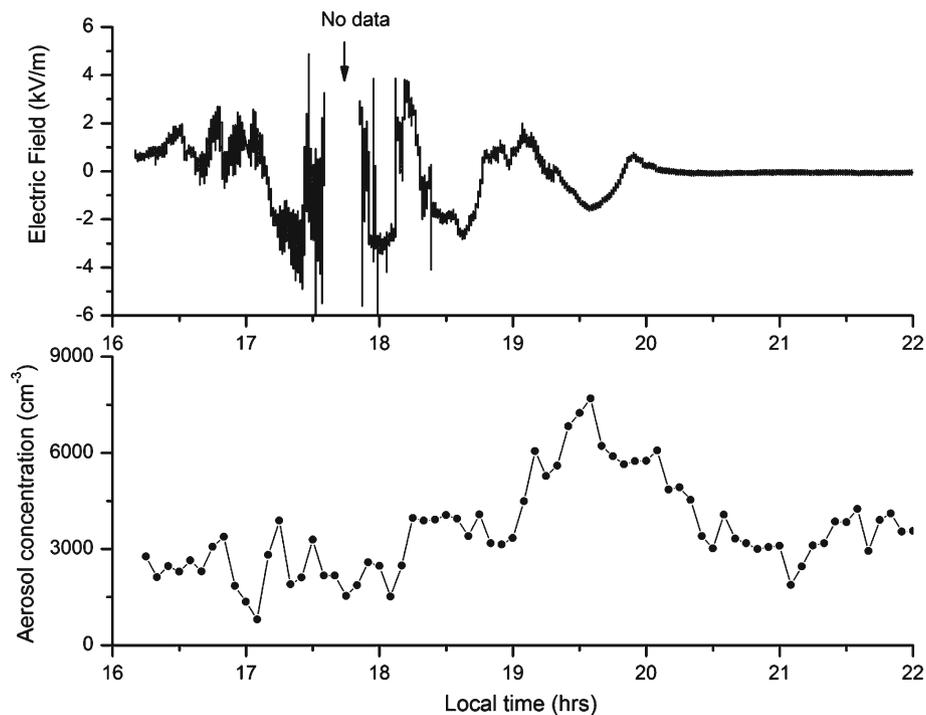


Figure 5. Variations of (a) electric field, and (b) concentration of aerosols of size below 50 nm diameter on May 22, 2008.

Heckman (1993) have shown that the deep clouds over tropical continents can be sufficiently electrified to produce corona current at earth's surface even though only some of them can produce light-

ning. Corona currents release unipolar ions into the atmosphere. Therefore, we propose that new particle formation by ion nucleation during process of corona discharges can contribute significantly to

the new particle formation over tropical continent. Our observations also suggest that unipolar ions released into the atmosphere by corona discharges may be more favourable for ion nucleation process and for subsequent growth of new particles.

As far as our knowledge, it is for the first time that new particle formation during dissipation stage of thunderstorm in unipolar ion environment has been reported. However, study of more number of such cases is required, not only to estimate the contribution of this process for global aerosol concentration but also to assess the role of corona produced ions in changing the aerosol size-distribution beneath a thunderstorm. We agree that lack of measurements of ions in different categories may cast doubt about our conclusions; however, increase in ion concentration during dissipation stage of thunderstorm is a well known phenomena (Standler and Winn 1979; Standler 1980; Livingston and Krider 1978; Krider and Musser 1982; Pawar and Kamra 2009) and it supports our conclusions.

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