Formation and Hygroscopic Growth Properties of Ultrafine Particles in College Station, Texas, in 2003

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During May of 2003, smoke from fires in the Yucatan Peninsula was transported across the Gulf of Mexico and into Texas where it caused significant enhancement in measured aerosol concentrations and reduced visibility. During this event, the formation and growth of aerosol particles has been observed by a differential mobility analyzer (DMA) / tandem differential mobility analyzer (TDMA) system to characterize the size distribution and size-resolved hygroscopicity of the aerosol. The most number concentration is by the particles smaller than 100 nm, but the integrated number concentrations for over 100 nm increased due to the aerosol growth. Hygroscopic growth factor increase from 1.2 to 1.4 for 25, 50, and 100 nm particles during the nucleating period. This distribution and the aerosol properties derived from the TDMA data were used to calculate the growth rate. Particle growth rates were in the range 1-12 nm/hr.

Key Words : Aerosol, Nucleation, Aerosol size distribution, Hygroscopicity, TDMA

1. Introduction

Aerosols can have significant impacts on local, regional, and even global radiative budgets. Despite a number of studies directed at quantifying the impact of aerosols, there is still considerable uncertainty regarding the radiative forcing associated with increasing concentrations[1]. Understanding these effects require detailed information on how aerosols form, enter, and transform in the atmosphere. Most important processes are the formation of new aerosol and subsequent growth to larger sizes. The characteristics of the nucleation episodes are studied in Makela et al.[2] and Vakeva et al.[3]. A number of recent studies have examined the nucleation events at a number of sites around the world[4]. They reviewed the formation and growth rates of ultrafine atmospheric particles and showed that typical particles growth rates are between 1 and 20 nm/hr in midlatitudes. Aerosol particles up to 100 nm in diameter are commonly called ultrafine particles.

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The aim of this study is to investigate the aerosol formation and characterize the hygroscopic growth properties. Growth rates are determined from measured nucleation mode size distributions.

2. Measurements

Intentionally set fires in agricultural regions on and near the Yucatan Peninsula generate elevated concentrations of particulates that impact aerosol loadings in the U.S. every spring. Biomass burning aerosol loadings in Texas during May of 2003 were significantly higher than during the previous four years. Aerosol size distributions and hygroscopic growth factors were measured in College Station (Texas A&M University), Texas between May 11 and 14 of 2003 after the biomass burning episode along with additional sites at which particulate matter measurements were made (Fig. 1). Additional data were recorded by the Texas Commission on Environmental Quality (TCEQ) at several sites throughout Texas.

3-day back trajectories ending in College Station, Texas were calculated during the episode for four different levels from 990 to 910 hPa using the National Centers for Environmental Prediction (NCEP) rean-
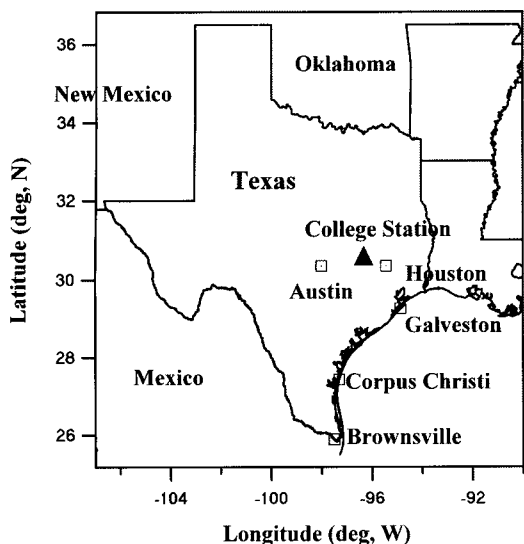
Fig. 1. Map of measurement location, College Station of southeast Texas.

alysis data. All of these trajectories confirmed that air was transported from the Yucatan to Texas. Fig. 2 shows two examples (11 and 12 of May 2003) of back trajectories, with both sets originating in the region influenced by biomass burning. Fig. 3 shows

traces of visibility and relative humidity recorded at Easterwood Airport in College Station. The rapid increase in visibility on May 11.5 marks the change of air mass.

Fig. 4 shows a timeline of PM2.5 mass concentration at several TCEQ sites from May 10 to 15. The maximum hourly PM2.5 concentration measured in several cities in Texas was about $40 \sim 70$ $\mu g/m^3$. Elevated concentrations were observed throughout the State from the southernmost tip in Brownsville, north through Corpus Christi, Galveston, and Austin. The biomass burning episode terminated rapidly with the passage of a front on May 11 noon. Throughout much of the period during which particulate matter concentration about 40 $\mu g/m^3$ was elevated during nucleation event just after biomass burning episode. The hygroscopic properties and size distribution of this aerosol in College Station, Texas were measured to characterize the nucleation event.

3. Size and hygroscopic growth factor measurements

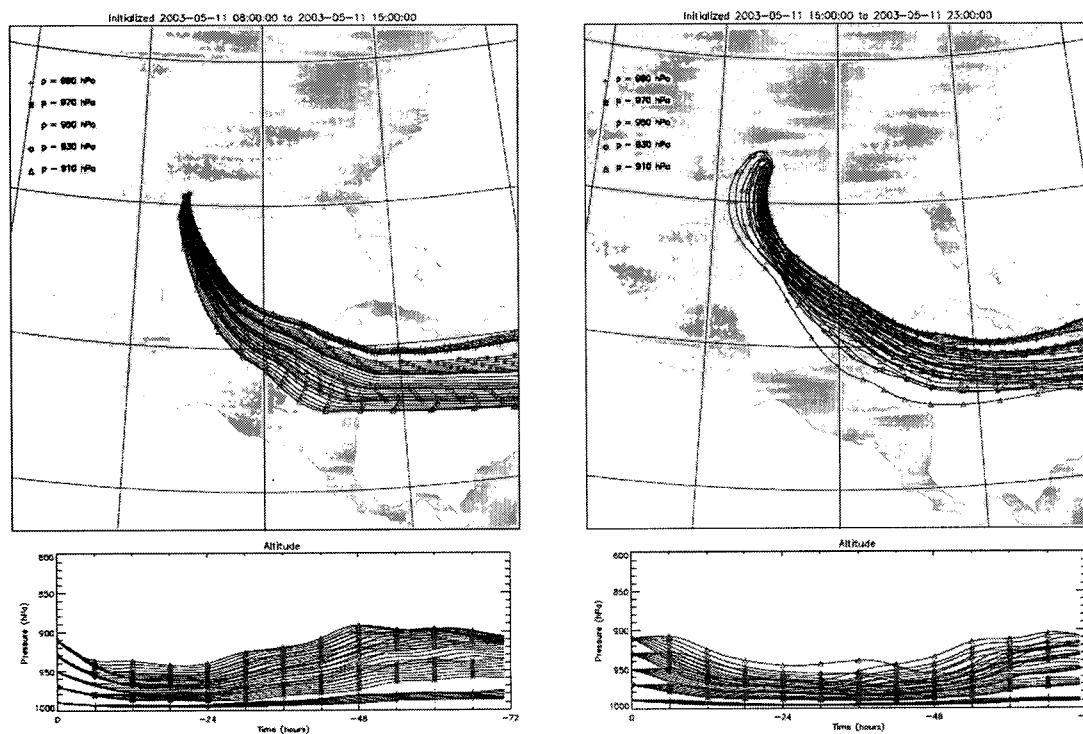Aerosol size distributions and hygroscopic growth factors were measured during the nucleation episode

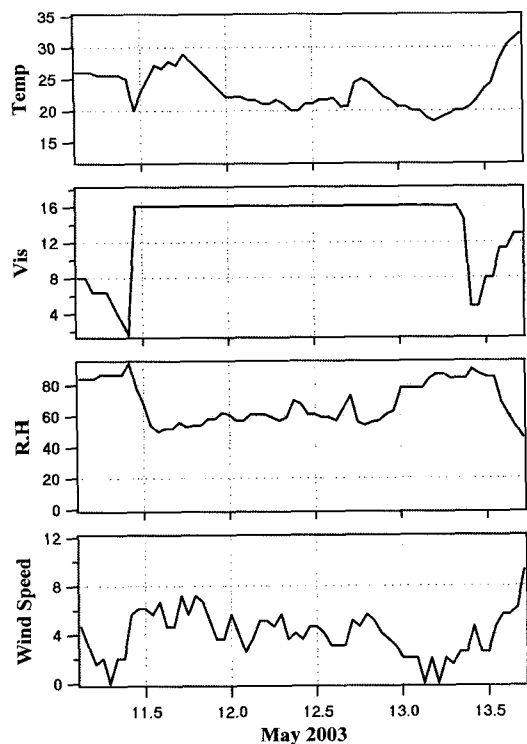Fig. 2. Example back trajectories ending in College Station, Texas on 11 and 12 May 2003.

Fig. 3. Time series of visibility (km), temperature (℃), wind speed (m/s), and relative humidity (%) recorded at Easterwood airport in College Station, Texas. Along the x-axis, 11.5 correspond to noon on May 11.
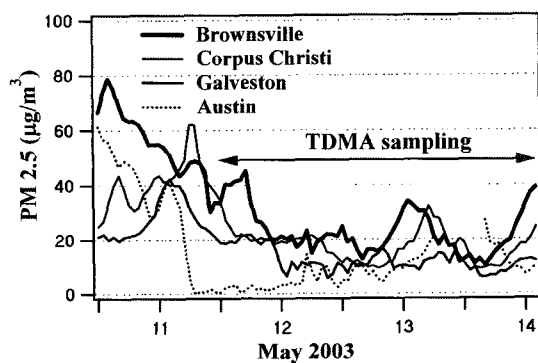

Fig. 4. Hourly PM2.5 concentrations at four TCEQ cities during the measurement period.

in College Station, TX between May 11 and 14 of 2003 after the biomass burning episode. The hygroscopic growth factor (D_p/D_p^*) is defined as the ratio of the hydrated diameter of the aerosol at elevated relative humidity (RH, 85 %), D_p to the initially separated particle diameter at low RH, D_p^*. A detail description of measurements is provided in the work of

Lee[5]. Throughout the study period, hygroscopic growth factors of particles having diameters of 25, 50, 100, 200, 300, 450, and 600 nm were measured approximately once every hour. Fig. 5 shows an example of number size distribution and hygroscopic growth factor distributions measured at 15:20 central daylight time (CDT) on May 11, 2003. The number size distribution ($dN/dlogD_p$) peaks between 40 and 50 nm with 18,000 cm^{-3}. This number size distribution of this nucleation aerosol is similar to those described by Stanier et al.[6]. The growth factor of the main mode were near the 1.0 at 600 nm, 1.3 at 25-100 nm, and 1.4 at 200-450 nm. The similar appearance of the distributions for all but the smallest and largest particles suggests the dominance of a single particle type. As shown in the figure, medium size distributions contained two hygroscopic modes. Examples of growth factor distributions shows that when aerosols are increased to larger size, they contains more modes and shapes are wider than single modes.
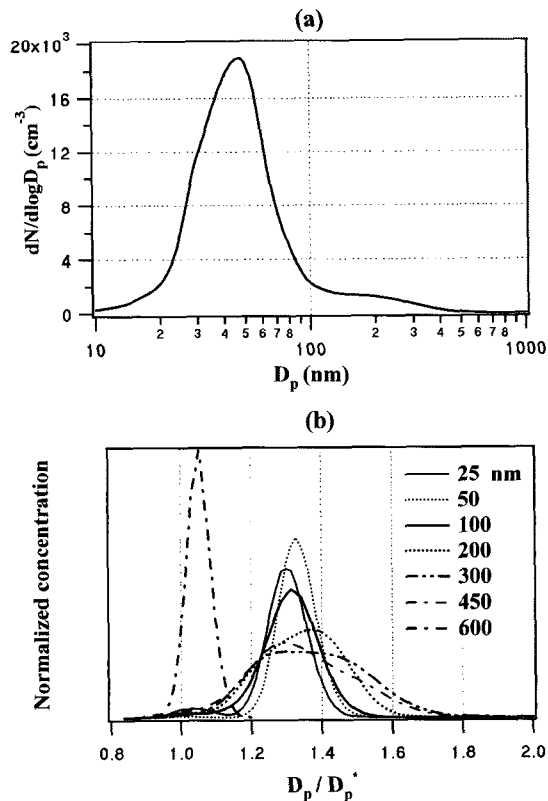

Fig. 5. Aerosol size distribution and fixed-RH hygroscopic growth factor distributions for 25 to 600 nm aerosol diameters measured at 15:20 CDT on 11 May 2003.

4. Discussions

4.1. Size distributions

The measured aerosol size distributions measured by DMA are shown in the contour plots in Fig. 6. Before midday newly formed particles below 10 nm enter the detectable size range, after which they grow at a rate of a few nm/hr, reaching sizes between about 50 and 100 nm by the evening. The total number concentration peaked between 11.5 and 12 May, while a shift in the size distribution towards greater particle size resulted in aerosol growth. The most number concentration is by the particles smaller than 100 nm. From the data sets it is possible to estimate the quantities size range with time evolution, after
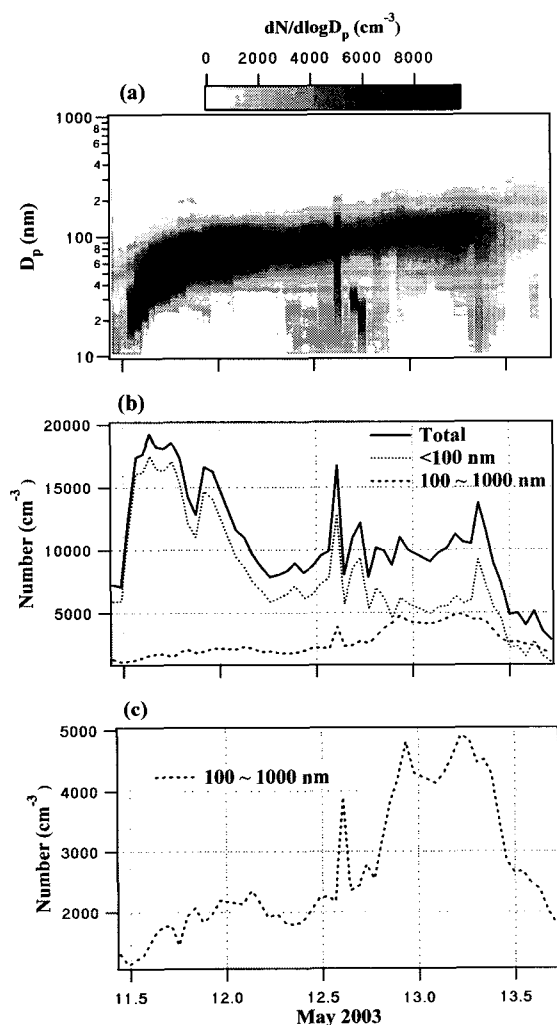

Fig. 6. (a) Size number concentration, (b) total number concentration, and (c) for in details.

which growth rate can obtained. Total number concentrations ranged from 2,500 to 18,000 cm^{-3} with an average of 10,000 cm^{-3} during the sampling period. The integrated number concentrations for over 100 nm increased due to the aerosol growth.

4.2. Aerosol hygroscopic growth

Studies of aerosol formation and growth require measurements of nucleation mode particles smaller than 20 nm. Particle formation and growth rates can be inferred from measurements of nanopartilce size distributions. Aerosol particles up to 50 nm in diameter are commonly called nanoparticles. The hygroscopicity of freshly nucleated particles can be measured with the TDMA, and it provides constraints on the composition of growing particles. When nanoparticles were humidified at high relative humidity, they became more hygroscopic during the growth period. Fig. 7 shows the size dependence of the hygroscopic behavior of the aerosol with diameters between 25 and 600 nm. During the nucleation episode hygroscopic growth increased rapidly at 25, 50, and 100 nm. The maxima in each of the contour plots in Fig. 7 corresponds to a significant concentration of particles during the nucleation event. But hygroscopic growth for particles 200 nm and larger are almost the same. The compositional heterogeneity of an aerosol is reflected in the spread in the hygroscopic growth factor distribution. The hygroscopic growth factor distributions for particles smaller than 100 nm were usually monomodal, whereas additional modes were frequently observed in the distributions for larger particles. The spread in the hygroscopic growth factor distribution with increasing particle size suggests a greater degree of heterogeneity with the larger particles considered[7].

4.3. Growth rate

Estimating the particle growth rate requires information on the time evolution of the mean diameter of the new particle mode. With this information, growth rate can be calculated from the formula GR $= \frac{\Delta D}{\Delta t}$, where D belongs to the mean range with time[4]. Particle diameter changes during nucleation and particle diameter growth rates during nucleation events are high. Typical particle growth rates are between 1 and 20 nm/hr in midlatitudes[4]. Fig. 8a shows mean size of nucleated aerosols versus time of day. Number
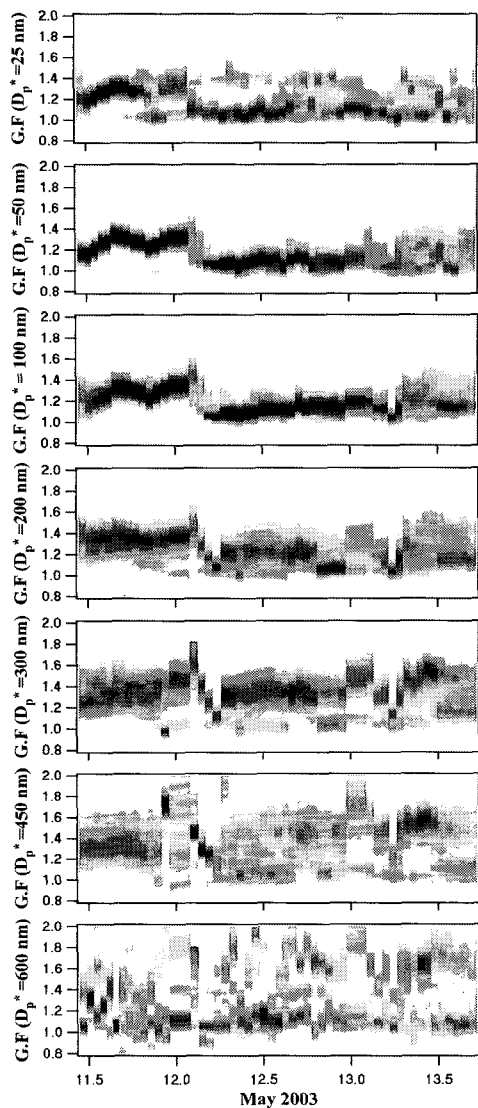
Fig. 7. Measured hygroscopic growth factor distributions for particles with diameters between 25 and 600 nm.

Fig. 8. (a) Number mean size and (b) growth rate versus time of day during a nucleation event in College Station on May 11-13, 2003.

mean diameter increased with time from 20 nm to 80 nm. The diameter growth rate is the slop of this line. Fig. 8b shows that particle growth rates are remarkably high during the 11 May 2003 with 11 nm/hr and decreased with time. This is explained by the increasing of the hygroscopic growth factor under the 100 nm size during the nucleating period.

5. Conclusions

A particle formation event was observed in the urban site. A DMA / TDMA system was used to characterize the size distribution and size-resolved hygroscopicity of
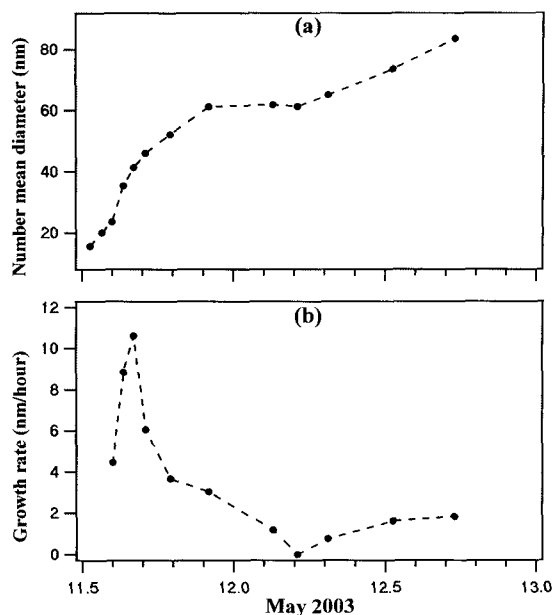
nucleating aerosol in College Station, Texas USA from May 11 ~ 15 of 2003. Measured size distributions and the aerosol hygroscopic growth properties derived from the TDMA data were used to calculate the concentration and growth rate. Particle growth rates were remarkably high during the 11 May 2003 and it also showed that hygroscopic growth factor increase during the nucleating period under the 100 nm size. During the nucleation event the growth factors of the nucleated particles increased from 1.2 to 1.4 for 25, 50, and 100 nm particles. Particle growth rates were between 1 and 11 nm/hr and showed the typical particle growth rates between 1 and 20 nm/hr in midlatitudes.

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