Observation of aerosol number size distribution and new particle formation at a mountainous site in Southeast China

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Abstract:

To quantify the physical/chemical properties, and the formation and growth processes of aerosol particles on mountainous regions in Southeast China, an intensive field campaign was conducted from April to July, 2008 on the top of Mt. Huang (1840 m above mean sea level). The average particle number concentration was $2.35 \times 10^3 \text{ cm}^{-3}$, and the ultrafine particles ($<0.1 \mu\text{m}$) represented 70.5% of the total particle number concentration. Excluding the accumulation mode particles, the average daytime particle number concentrations were prominently higher than those measured at nighttime, suggesting there was a diurnal pattern of changes between planetary boundary layer and free troposphere air. The aerosol spectra were classified into two categories: the first category (FCS) exhibited a clear diurnal cycle, with relatively higher number concentration ($3.19 \times 10^3 \text{ cm}^{-3}$), smaller sizes and air masses from the inland; the second category (SCS) presented less obvious diurnal cycle, with lower number concentration ($1.88 \times 10^3 \text{ cm}^{-3}$), larger sizes and air masses from coastal regions. Air mass sources, weather conditions, and new particle formation (NPF) events were responsible for the differences of these two particle spectra. Six NPF events were identified, which usually began at 10:00-11:00 LT, with the estimated formation rate J_{10} in the range of $0.09\text{--}0.30 \text{ cm}^{-3} \text{ s}^{-1}$ and the growth rate at $1.42\text{--}4.53 \text{ nm h}^{-1}$. Wind speed, sulfur dioxide and ozone concentrations were higher on NPF days than those on non-NPF days, whereas temperature, relative humidity, concentrations of nitrogen oxide and carbonic oxide were lower on NPF days. Sulfur dioxide and ozone might be main potentially precursor gases for those NPF events. The NPF events at Mt. Huang corresponded closely to a southwest winds. These results are useful for improving our understanding of the main factors controlling the variation of aerosol size distribution and NPF events in this region.

Keywords: Aerosols; Size distribution; High mountain; Southeast China; New particle formation

1. Introduction

Atmospheric aerosol particles can affect the Earth's energy budget through directly scattering and absorbing solar radiation (Charlson et al., 1992; Heal et al., 2012), and indirectly impacting the cloud albedo and precipitation by serving as cloud condensation nuclei (CCN) or ice nuclei (IN) (Twomey, 1974; Albrecht, 1989; Lebo and Feingold, 2014). However, the uncertainties of these effects are still considerable (Solomon et al., 2007), which require temporally and spatially representative datasets about chemical/physical properties of aerosol particles. One of the key physical parameters is the number size distribution in the submicron range (Kumar et al., 2014). In fact, submicron particles make a significant contribution to particle number concentration (Morawska et al., 1998), and studies also show evidences of their adverse effects on human health (Charlson et al., 1992; Rodriguez et al., 2005). Therefore, the knowledge of particle size distribution especially for submicron particles is crucial in avoiding human exposure, estimating climate effects, and designing controlling strategies (Charlson et al., 1992; Kumar et al., 2014).

New particle formation (NPF), associated with a rapid burst of nucleation mode particles, results in increasing the total number concentration of submicron particles (Twomey, 1976). Observations revealed that the NPF events and subsequent growth are universal in the atmosphere. The formation rates of 3 nm particles often range from 0.01 to 10 cm⁻³ s⁻¹ in the boundary layer and higher (up to 100 cm⁻³ s⁻¹) in urban areas. However, the formation rates as high as 10⁴-10⁵ cm⁻³ s⁻¹ in coastal areas and industrial plumes were also reported (Kulmala et al., 2004). Typical particle growth rates vary from 1 to 20 nm h⁻¹ in mid-latitudes depending on the temperature and the availability of condensable vapors, such as sulfuric acid and possibly other organic compounds with low saturation vapor pressure formed via photochemical reactions (Kulmala et al., 2004; Zhang et al., 2004). Moreover, larger pre-existing particle concentrations suppress the formation of new particles by consuming condensable vapors concentrations and accelerating the coagulation scavenging of newly nucleated particles (Kerminen and Kumala, 2002; Kulmala et al., 2004).

Field measurements of particle size distribution in the submicron range size and NPF processes have been conducted in the free troposphere, Arctic, boreal forest, marine, urban and rural environments (Zaizen et al., 1996; Reus et al., 2000; Komppula et al., 2003; Stanier et al., 2004; Dal Maso et al., 2005; Rodriguez et al., 2005; Lee et al., 2008). Observations of particle size distribution and NPF events were also carried out in several megacities and relatively polluted

regions in recent years in China, such as Beijing (Yue et al., 2009), Pearl River Delta (PRD) (Liu et al., 2008; Chen et al., 2016), and Yangtze River Delta (YRD) (Gao et al., 2009; Wang et al., 2014; Qi et al., 2015). Those studies show that the number concentrations of accumulation mode particles are relatively high in urban areas, and the NPF events are much more frequently occurred in more remote locations (Kulmala et al., 2004; Wu et al., 2008).

The atmosphere in mountainous areas is usually controlled by lower temperature, higher relative humidity, and more intense solar radiation conditions (Seinfeld et al., 2004; Li et al., 2011). Tropospheric aerosols over mountains are often impacted by long-range transport (Li et al., 2011; Wang et al., 2012). The size distribution and their formation and growth of submicron particles have been characterized at some high altitude stations (Venzac et al., 1995; Nyeki et al., 1998; Weingartner et al., 1999; Nishita et al., 2008; Rodriguez et al., 2009; Kanehiro et al., 2011; Guo et al., 2012), and in China, a few studies were conducted at Mt. Waliguan (3816 m), remote western China (Kivekäs et al., 2009), Mt. Tai (1534 m), east China (Zhang et al., 2014), and Mt. Hua (2060 m), central China (Wang et al., 2012). In southeast China, only two studies were performed on Mt. Huang to characterize the aerosol number size distribution and NPF events, but both were conducted at the foot of Mt. Huang in the autumn (Wang et al., 2014; Hao et al., 2015). Wang et al. (2014) laid emphasis on the effects of air masses on aerosol spectral distribution and the characteristics of NPF. The various characteristics and influential factors on aerosol size distribution, the favorable conditions and potential gaseous compounds participating in NPF and growth have remained still unclear at the top of Mt. Huang. Therefore, it is essential to enrich the database of particle size distribution and NPF observations at this high altitude site.

An intensive field campaign to measure the concentration, size distribution, chemical composition of aerosol particles (10-10000 nm) and related quantities (including trace gases and meteorological parameters) was conducted at the top of Mt. Huang (1840 m above mean sea level) in southeast China (Fig. 1) from April to July in 2008. The aims of this paper are two folds: Firstly, to characterize the particle number concentration, size distribution and new particle formation events; Secondly, to explore the possible causes for different distribution characteristics, favorable meteorological conditions and potential precursor gases participating in the nucleation and growth of particles.

2. The experiments and methods for data analysis

2.1. The background of the experimental site

The field experiments were conducted at the Bright Summit (Peak Guang Ming Ding, 118°09' E, 30°10' N, 1840 m a.s.l.), one of the highest peaks of Mt. Huang, which is located in the south of Anhui province, southeast China (Fig.1). This site is distant from the major cities of

Anhui province and further located about 186 km away from Hangzhou, the nearest megacity. Mt. Huang is not directly influenced by industrial emissions and is generally regarded as a clean background site (Hao et al., 2015). And also, due to its special geographical location and complex atmospheric conditions, Mt. Huang is often covered with clouds or fogs, and is regarded as a natural laboratory for studying aerosol-cloud/fog interactions.

(Fig.1)

2.2. Instruments and data analysis

From April to July, 2008, a Wide-range Particle Spectrometer (WPSTM, MSP Co., USA model 1000XP) was employed to measure aerosol number concentration and size distribution in the range of 10-10000 nm. It is a high-resolution aerosol spectrometer that combines the principles of differential mobility analysis (DMA, MSP Co., USA), condensation particle counting (CPC, MSP Co., USA), and laser light scattering (LPS, MSP Co., USA).

The DMA in the WPSTM has a cylindrical geometry with an annular space for the laminar aerosol and sheath air flows. The CPC is of the thermal diffusion type, with a saturator maintained at 35 °C. It has a dual reservoir design to prevent the working fluid from being contaminated by condensed sampling-air humidity. The LPS is a single-particle, wide-angle optical sensor. The DMA and the CPC can measure particles in the size range of 10-500 nm and can be setup to the maximum of 96 channels. The LPS covers the range of 350-10000 nm and is divided into 24 additional channels. Here we chose the sample mode with 48 channels in DMA and 24 in LPS. Thus it took approximately 8 min for one complete scanning of the entire size range with scanning period of 5 s for each channel. Ambient ozone (O₃) was monitored using a UV photometry analyzer (TEI model 49i, TSI, USA), carbon monoxide (CO) using a gas filter correlation analyzer (TEI Model 48i, TSI, USA), sulfur dioxide (SO₂) by a pulsed fluorescence analyzer (TEI model 43i, TSI, USA), and nitrogen oxides (NO_x) with a modified Chemiluminescence NO-NO₂-NO_x analyzer (TEI Model 42i, TSI, USA). The calibrations of those sampling instruments were showed in Table 1, and more detailed information was introduced by Liu et al. (2010) and Wang et al. (2013).

(Table 1)

The quality of the data given by WPS was determined by the operation procedures provided by the manufacture. Due to April to August is the foggy and cloudy season on Mt. Huang, the high humidity and fog/cloud water restrained the proper performance of the sampling instruments. So,

the samples were abandoned when measurements lasted for less than consecutive 12 hours a day. In addition, the data collected in 30 mins after the reboot of the instruments were also rejected. Similarly, the quality assurance and control procedures were also performed for trace gas data, calibrating the observed data and excluding the data with large error due to machine switch and equipment failure. Meanwhile, meteorological parameters including the air temperature, relative humidity, precipitation, wind speed and direction were provided by the automatic meteorological station at Bright Summit after quality control. A total of 2849 samples and 16 effective observation days (from April 22 to May 7) were used in the following analysis.

The following parameters were also used to describe the aerosol size distribution, including the geometric mean diameter (GMD) and the geometrical standard deviation (σ). Which was defined D_g (GMD) as:

$$D_g = \exp\left(\frac{\sum_i N_i \ln(D_i)}{\sum_i N_i}\right) \quad (1)$$

Where, N_i is the number concentration for a specific bin i , and D_i is the average diameter of bin i .

3. Results and discussions

3.1. General characteristics of particle number concentration and size distribution

Fig.2 presented the time series of particle number size distribution, total number concentration, temperature, RH, wind, air pressure, and precipitation intensity measured during study period. During the campaign (from April 22 to May 7), there were a series of sunny days with pronounced diurnal variation of temperature and relative humidity (RH), and several cloudy days were also occurred with continued higher RH. The mean temperature for the campaign was 11.0 ± 4.6 °C. The average RH was 71 ± 25 %, and the accumulated rain (26 mm) was relatively low during this period of the time. The main wind direction was southwest with an average speed about 6.0 ± 3.6 m/s. The atmospheric pressure slightly changed as well. In summary, the weather conditions during the campaign were characterized by moderate instability.

(Fig.2)

Table 2 gave statistical results of particle number concentration in different size ranges. The average particle number concentration was $2.35 \times 10^3 \text{ cm}^{-3}$, and particles in size range of 0.01-0.5 μm represented 98.7% of the total particle number concentration. On average, 70.5% of the total particle count was found in the size range of 0.01-0.1 μm , revealing abundant ultrafine particles ($<0.1 \mu\text{m}$) in this region. Particle number concentrations in both 0.01-0.1 μm and 0.01-0.5 μm were an order of magnitude lower than observational results obtained in urban/suburban areas

(Ruuskanen et al., 2001; Stanier et al., 2004; Qian et al., 2008; Gao et al., 2009; Zhai et al., 2011). However, the best measurements to compare our results with were those conducted at mountainous sites. As shown in Table 3, the general level of particle number concentration in the size range of 0.01-0.5 μm here was similar to that measured at those foreign mountainous regions, but universally lower than that at Mt. Tai and Mt. Waliguan in China, especially for ultrafine particles. This indicated that the Mt. Huang was relatively less polluted and this site could be considered as a clean continental area. Furthermore, aerosol number size distribution was widely divided into a nucleation mode (3-20 nm), representative of newly formed particles, an Aitken mode (20-90 nm), and an accumulation mode (90-1000 nm) representing more aged particles (Kulmala et al., 2004). Fig.2 showed the consecutive evolution of the size distribution and number concentration in size range of 0.01-10 μm . The particle counts showed a large temporal variability with spikes higher than 6800 cm^{-3} on several occasions (24, 25, 26 April and 5, 6 May) exceeding the average number concentration by a factor of three. The sharp increases in nucleation mode and Aitken mode particles dominated most of the particle bursts, which mainly occurred on sunny days due to nucleation processes. Nucleation mode particles were hardly observed on cloudy and rainy days (29, 30 April and 2, 3, 4 May).

(Table 2)

(Table 3)

The average diurnal variations of particle number concentration in the size range of 10-20 nm (nucleation mode), 20-90 nm (Aitken mode), 90-1000 nm (accumulation mode), and 10-10000 nm were showed in Fig.3. We could find that the nucleation mode particle number concentration rapidly increased around 9:00 and kept relative high values until 20:00. Such pattern was not found clearly for the accumulation mode. The diurnal variation of total particle number concentration was very similar to that of Aitken mode. The average total number concentration peaked around noon local time, and an additional peak was also obvious in the late afternoon (around 19:00). To show the diurnal variation of particle spectra, we divided the average number size distribution into two classes: daytime spectrum (9:00-20:00, LT) and nighttime spectrum (21:00-8:00, LT) (Fig.4). Results showed that the overall particle number size distribution appeared obvious diurnal variation, with a bimodal and a single modal distribution prevailed during the day and night, respectively. It was dominated by smaller Aitken mode particles (with a mode size of 28 nm) in the daytime and larger Aitken mode particles (with a mode size of 58 nm) at nighttime. Furthermore, average and median particle number concentration of each mode for daytime and nighttime values were calculated in Table 4. The total particle number concentration

was higher during daytime than nighttime, when the ratios of both average and median values were above 1.5. Similar behaviors were also found in both nucleation mode and Aitken mode. These findings indicated that there was a diurnal pattern of changes between airs coming from the planetary boundary layer (PBL) during the daytime and from the free troposphere (FT) during the nighttime at the top of Mt. Huang, coincident with other studies which were also conducted at mountainous sites (Nyeki et al., 1998; Nishita et al., 2008; Boulon et al., 2011). It was noteworthy that the ratio was higher than 10 for nucleation mode, suggesting frequent formation of nucleation mode particles during daytime in the study area.

(Fig.3)

(Fig.4)

(Table 4)

The continuous spectra of aerosol particle number concentration can more intuitively show the distinct characteristics of aerosol spectra under different weather conditions. Based on the weather conditions, the continuous distribution spectra could be divided into two categories: the first category (hereafter: FCS) occurred mainly on sunny days from April 24 to 28 and May 5 to 6, 2008, respectively; the second category occurred mainly on cloudy days from April 29 to May 4 (hereafter: SCS). The FCS showed a significant diurnal variation, especially for particles in Aitken mode (Fig.2). The GMD of FCS reached maxima in the morning, then plummeted to the minimum, then slowly rose to maximum before dawn in the next day. Such characteristics were also observed in boreal forests (Hämeri et al., 2003). However, the diurnal variation in the SCS was very weak and the Aitken mode particles concentrations were relatively low (Fig.2). The GMD of the SCS was fairly constant, though it remained a high value about 110 nm in most time but suddenly occurred in a smaller value sometimes. Even so, the GMDs were inversely proportional to the number concentrations in both FCS and SCS, that was, when the GMD was small, the aerosol number concentration was high, and vice versa. It indicated that the increase of particle number concentration mainly caused by the increasing small particles.

(Fig.5)

The average number size distribution of FCS (Fig. 5) was characterized as a single modal distribution, with a broad Aitken mode peaked at 33 nm and a particle count of more than 3000 cm^{-3} and ultrafine particles ($<0.1\mu\text{m}$) represented 75.4% of the total number concentration ($3.19 \times 10^3 \text{ cm}^{-3}$) (Table 2). In contrast, the total number concentration of SCS ($1.88 \times 10^3 \text{ cm}^{-3}$) was

much lower than that of FCS (Table 2). The SCS number size distribution was characterized as bimodal, with a larger peak at 102 nm (accumulation mode) and an inferior peak at 18 nm (nucleation mode) (Fig. 5). Particles in size range 0.1-0.5 μm accounted for 37.0% of the total particle number concentration in SCS, much higher than that in FCS (accounting for 24.0%) (Table 2). It suggested that the FCS was dominated by smaller sizes (Aitken mode) particles and higher number concentration, while the SCS was characterized by higher concentration of larger sizes (accumulation mode) particles and lower number concentration.

Combined with the category for daytime and nighttime due to different aerosol spectra characteristics, the aerosol number size distribution of FCS and SCS could be further divided into daytime spectrum and the night spectrum (Fig.6). The key point from Fig.6 implied that the characteristics of number size distribution mainly depend on other factors (such as aerosol sources and weather conditions) not just on daily timescales (daytime or nighttime) to some extent.

(Fig.6)

3.2. The possible causes for the characteristics of number size distribution

The air mass analysis is one of the most common methods to analyze the different aerosol characteristics and sources. Using the HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model, we simulated the air mass trajectories from April 24 to May 6, 2008 at Bright Summit. Considering the vertical convection transport, 48-h back trajectories were modeled at the height of 1400 m and 800 m above ground level (corresponding to 1840 m and 1240 m above mean sea level respectively) (Fig. 7 and Fig. 8). The upper air masses were transported from the northwest direction and the highest altitude was over 4000 m, which came from the long-distance transport from Asian mainland for both FCS and SCS. However, the FCS and SCS were opposite in lower air mass. The lower air masses were transported from the continental southwest at relatively fast speeds over long paths in central troposphere about 1000 m in FCS, while that in SCS mainly originated from the coastal south to southeast and passed slowly along short paths. The continental air masses in FCS contained lower humidity and more pollutants. The coastal air masses in SCS contained abundant water vapor and a part of particles have been cleaned by precipitation occurred in this time-interval (Fig. 2). As stated by Putaud et al. (2003) and Kim et al. (1995), the higher number concentration and dominant Aitken mode particles often occurred in mainland air masses, while the lower number concentration and dominant accumulate mode particles often occurred under oceanic influence. Same results were also observed in Shenzhen (Shi et al., 2009). Thus, it indicated that the air mass sources had great impact on the characteristics of particle spectra, resulting in the continental spectral characteristics of FCS and

the maritime spectral characteristics of SCS in this study. However, our results were different from the observations performed at the foot of Mt. Huang, in the autumn 2011 (Wang et al., 2014), which showed that oceanic air masses transported ultrafine particles but inland air masses carried accumulation mode particles. There were some factors might be responsible for such differences, including the time and height of simulation, the specific sources and the residence times of air masses, and so on. Meanwhile, the NPF events occurred in FCS in our study also made a large contribution to Aiken mode particles.

(Fig.7)

(Fig.8)

As stated above, the FCS mainly occurred on sunny days, while the SCS mainly occurred on cloudy days. Thus, the meteorological factors of different weather conditions played an important role on the aerosol size distribution. In Fig.9, it could be found that the variation of GMD agreed well with RH in FCS. The GMD of the aerosol particles was small when the RH was low, while the GMD increased with the increasing RH. This implied that the GMD of aerosol particles increased via hygroscopic growth in FCS. Meanwhile, there was an inverse correlation between the GMD and temperature in FCS, with the plunge in the GMD just happened in the period of rising temperature. The influence of temperature on the GMD might have three aspects: (1) with the increase of the temperature, the RH was dropping, limiting the hygroscopic growth; (2) particles which had already gone through hygroscopic growth in low-level were lifted caused by a strong thermal convection, which could not be observed by the instruments; (3) particles which had already gone through hygroscopic growth would lose moisture due to the increase of temperature (solar radiation), and led to a phenomenon of reverse Köhler curves.

(Fig.9)

However, the relationships between the GMD and temperature or RH in SCS were not clear as that for FCS (Fig.10). The RH was relatively high in days of SCS, and precipitation occurred on 4 May. Because the atmospheric structure was stable, the vertical movement caused by the thermal convection was weak, depressing the spread of polluted air masses. Meanwhile, the water vapor was sufficient but the large aerosol particles were dominant, so that formation of new particles no longer the main affecting factors for the aerosol spectrum change in SCS. The GMD suddenly dropped on 4 May because of precipitation (precipitation could remove large particles). Thus, the rangeability of GMD was relatively stable but discontinuous, and the cause of the

suddenly drop of GMD might be explained by the variation of other parameter such as wind speed (WS) rather than temperature and RH in SCS.

(Fig.10)

From Fig.11 (a), we could see that when the WS was high, the GMD was large and its variation was stable. When the WS was lower than 2.5 m/s, the GMD suddenly drop. This phenomenon could be resulted from the increasing small particles occurred in low WS, such as nucleation mode particles. Fig. 11 (b) showed clearly that high levels of nucleation mode particles (10 -20 nm) appeared only in the time with low WS. This is caused by the low WS with restrictions on the diffusion/dilution of the newly generated particles and the condensable vapors which contributes to the formation of nucleation mode particles (Hao et al., 2015). In general, the rapidly increase of nucleation mode particles under low WS resulted in the decrease of the GMD in SCS.

(Fig.11)

3.3. New particle formation and growth events

An NPF event was defined as the appearance of a distinct new mode in the nucleation mode size range for a prominent period of time (hours) followed by the growth of newly formed particles to larger sizes (Dal Maso et al., 2005). In this study, an NPF event was defined if particle number concentration within the first nine WPS size bin (diameter midpoint of 10.41, 11.3, 12.25, 13.3, 14.43, 15.65, 16.98, 18.42, or 19.99 nm) bumped up with an increment of particle number concentration of size range 10-20 nm (N_{10-20}) larger than $\sim 10^2 \text{ cm}^{-3}$. And this criterion of number concentration defined the initiation time of the NPF event on each day. The growth rate (GR) was calculated using the formula:

$$\text{GR} = \frac{\Delta D_m}{\Delta t} \quad (2)$$

Where, D_m belongs to the size range $[D, D_{\max}]$ and the most frequently used selection for Δt is the duration of the particle formation event (Kulmala et al., 2004; Dal Maso et al., 2005). The unit of growth rates is nm h^{-1} . 10 nm particle formation rate (FR, J_{10}) was calculated for all NPF days by the following formula (Kulmala et al., 2012), taking into account the coagulation losses and growth losses:

$$J_{10} = \frac{dN_{10-20}}{dt} + \text{Coag} S_{10-20} N_{10-20} + \frac{1}{10\text{nm}} \text{GR}_{10-20} N_{10-20}$$

Where, N_{10-20} is the particle number concentration in the size range of 10-20 nm, $\text{Coag}S_{10-20}$ is the sink of 10-20 nm particles by coagulation and GR is their growth rate according to Kulmala et al. (2012). The unit of formation rates is $\text{cm}^{-3} \text{s}^{-1}$.

To evaluate the drivers for NPF at Mt. Huang, various atmospheric variables were analyzed to compare between NPF days and non-NPF days, including concentrations of trace gases (SO_2 , NO_x , CO and O_3), and meteorological parameters (RH, temperature, wind). Data from the 3 h prior to the initiation time of each event and to the average initiation time (i.e. 8:00-10:00 LT) were taken into account on NPF event days and non-NPF event days respectively.

A total of 6 NPF events (frequency of 37.5%) were found during the observation period (Table 5). All NPF events appeared in FCS which mainly occurred on sunny days, and these events were initiated at about 10:00-11:00 when the solar radiation rapidly increased and the boundary layer developed, except for one case in 5 May began in afternoon (13:00). The observations are consistent with other studies (Stanier et al., 2004; Hamed et al., 2007; Guo et al., 2012). Given it takes at least a couple of hours for newly formed particles to grow into the minimum detectable size of our aerosol instruments (10 nm), the real initiation time of these NPF events could be earlier than the observed initiation time (Kulmala et al., 2004; Kivekäs et al., 2009). Meanwhile, the averaged end time was 20:38 when the boundary layer was stable and the solar radiation became weak, implying that the NPF and growth process was closely related to the solar radiation and the development of boundary layer (Boulon et al., 2010, 2011). The average duration was 9.8 hours with a minimum of 6.2 hours and a maximum of 12.5 hours. According to Hamed et al. (2007), NPF event having an intensive and distinct formation of small particles followed by continuous growth to larger sizes with an average of about eight hours could be classified into the class I. Thus, most of the NPF events in our study could be classified as class I, indicating that the NPF events on the top of Mt. Huang had uniform characteristics.

Compared with the studies of NPF events observed at the foot of Mt. Huang (Wang et al., 2014; Hao et al., 2015), the average duration of NPF events in this study was longer. The differences in processes involved in the NPF events at different altitudes might explain this difference: (1) the transportation due to the PBL development and the strong upslope wind which occur during daytime brings condensable vapors to the mountain-top (Venzac et al., 1995; Nishita et al., 2008); (2) the lower temperatures at higher altitudes increase the saturation ratio of condensable species (Boulon et al., 2011); (3) the pre-existing particle concentration and hence the condensational sink (CS) decrease with the altitude. These factors were evidenced by the fact that the nucleation frequency was higher at the top of Mt. Huang (37.5%) than at the foot of Mt. Huang (18.2%) (Hao et al., 2015). Those observations suggested that the nucleation and subsequently

growth process could have a large vertical extent, and the nucleation process is clearly enhanced at the higher altitude station, which was similar to the results obtained by Boulon et al. (2011) at two different rural altitude sites in central France.

However, the characteristics of each NPF event were not the same. The particle formation rates ranged from 0.09 to 0.30 $\text{cm}^{-3} \text{s}^{-1}$ with an average value of 0.23 $\text{cm}^{-3} \text{s}^{-1}$, lower than the results at the foot of Mt. Huang (Wang et al., 2014) and other results obtained on mountains-top sites (Boulon et al., 2010, 2011; Guo et al., 2012; Hallar et al., 2011; Shen et al., 2016). The particle growth rates varied from 1.42 to 4.53 nm h^{-1} with an average value of 2.90 nm h^{-1} . This average GR was similar to those in background sites and mountains-tops (Weingartner et al., 1999; Nishita et al., 2008; Kulmala et al., 2004; Guo et al., 2012; Shen et al., 2016), but lower than those of polluted areas (Yue et al., 2009; Wang et al., 2014; Qi et al., 2015). It suggested that the NPF event is a local phenomenon, which may be affected by the particle concentration in background atmosphere, local emissions and long-range transports (Hamed et al., 2007). Meanwhile, the dominant air masses of NPF events were from the southwest, where the humidity was relatively low.

(Table 5)

(Fig.12)

To present a visible picture of NPF and growth process, one NPF event occurred on 6 May was selected for the case study (Fig.12). NPF event on 6 May was obviously observed after raining on 4 May, according the fact that NPF events often appear after a sharp decrease in concentration of pre-existing particles (Liu et al., 2008; Guo et al., 2012). The NPF started when the wind speed decreased. The wind speed then sustained fairly low (about 2 m s^{-1}), which benefited the stay of gaseous pollutants and avoided the new particles from being blew away. The RH maintained below 90% all day and fell to less than 55% when the NPF event occurred. The lower RH is favorable to the condensation of the gaseous pollutants which participating in the nucleation and growth of particles (Wu et al., 2007; Yao et al., 2010). Meanwhile, slow increase in NO_x and O_3 , and obvious increase in SO_2 were observed, accompanied with the rise of the PBL height (Wang and Miao, 2013). As shown in Fig.12, the nucleation mode particle number concentration (N_{nuc}) was basically less than 50 cm^{-3} during 00:00-10:00, showing a really low N_{nuc} in the background atmosphere before the NPF event. The N_{nuc} began to rise at about 10:30 LT and increased rapidly from $\sim 2.89 \times 10^2 \text{ cm}^{-3}$ to $\sim 7.67 \times 10^2 \text{ cm}^{-3}$ at noon with a formation rate (J_{10}) of 0.009 $\text{cm}^{-3} \text{s}^{-1}$. With a significant GR of 4.53 nm h^{-1} , the number concentration of Aitken mode particles (N_{Ai}) began to increase at 11:00, reached to the maximum 2 hours after and maintained

at about 5000 cm^{-3} in the following 1 hour, which was due to the collision /adsorption and the hygroscopic growth of the newly formed particles (Wang et al., 2014). The particle number concentration in accumulation mode range (N_{acc}) started to grow then, but its increasing rate was lower than that of N_{Ait} . After 13:00, the GR decreased with the decreasing SO_2 . Additionally, correlation coefficient of N_{nuc} with SO_2 for the duration of this NPF event was -0.35 ($P < 0.05$) based on the one-hour averages, indicating that the depletion in SO_2 was responsible for the nucleation and growth of new particles. The wind direction turned from southwest to south at 14:00 but the wind speed kept fairly low for the whole time. However, there were obviously high concentrations of aerosol particles occurred on 17:00, dominated with Aitken mode and accumulate mode particles. At the moment, the RH and concentrations of trace gases significantly increased, while the temperature decreased. This was likely caused by the transportation due to the advection current or upslope wind. After 23:00, the total particle number concentration decreased gradually to the background concentration at about 24:00.

(Table 6)

To investigate the favorable conditions for NPF events, the concentrations of trace gases (SO_2 , NO_x , CO and O_3) and meteorological parameters (RH, T and wind) between NPF days and non-NPF days were compared (Table 6). Data from the 3 h prior to the initiation time of each event were taken into account on NPF event days, while data from 8:00-10:00 LT were considered on non-NPF event days. As shown in Table 6, the average temperature was slightly lower in NPF events than that in non-NPF events, similar to other observations (Boy and Kulmala, 2002; Komppula et al., 2003), due to the lower temperature at the initiation times of NPF events favored to the nucleation of sulfuric acid with water vapor (Guo et al., 2012). The average RH on NPF event days was lower than that on non-NPF event days (Table 6), revealing that the NPF events tended to occur on low RH. Two possible reasons could explain this phenomenon: (1) less clouds on lower RH days generally, resulting in the enhancement of downward solar radiation flux and OH radicals production subsequently, which contributed to the condensation of vapors (Hamed et al., 2007); (2) the hygroscopic growth of the pre-existing particles in high RH conditions could increase the condensational sink, which inhibited the nucleation of particles (Hamed et al., 2011). The NPF events at Mt. Huang correspond more closely to a westerly/southwesterly wind direction, while south/southeast winds were mainly observed on non-NPF event days (Table 6 and Fig.2). It is consistent with the back trajectory results which showed that the lower air masses were transported frequently from southwest, representative of the continental air masses on NPF days, while south to southeast air masses, representing the coastal air masses on non-NPF days. The

wind speeds on NPF days were slightly higher than that on non-NPF days (Table 6). Zhang et al. (2011) showed that strong winds were often observed before NPF events, due to high wind speeds are responsible for the effective dilution and diffusion of the aged aerosol in the atmosphere, which contributes to the formation of new particles.

For trace gases, it was found that the average concentrations of NO_x and CO (1.8 ± 1.0 ppbv and 261.5 ± 217.1 ppbv) were lower on NPF event days than those (2.9 ± 1.4 ppbv and 539.4 ± 184.0 ppbv) on non-NPF event days (Table 6). This could be partly due to the stronger solar radiation on NPF event days. The enhancement of solar radiation would accelerate the depletion of NO_x and CO concentrations through the photochemical reactions. However, the O_3 and SO_2 values (70.9 ± 6.5 ppbv and 5.6 ± 3.6 ppbv) 3 h prior to the NPF initiation were higher on event days than those values (60.8 ± 13.4 ppbv and 3.8 ± 3.0 ppbv) on non-NPF event days (Table 6), which is in line with other studies (Hamed et al., 2007). Guo et al. (2012) indicated that O_3 could be the precursor of NPF as it contributes to the formation of condensable vapors by directly reacting with VOCs, and indirectly generating OH and HO_2 radicals upon photolysis. Studies also showed that SO_2 is required for NPF events to produce sulfuric acid, via reaction with oxidant (hydroxyl and nitrate radicals, ozone), which is an important vapor precursor for the nucleation and growth process of stable nanometer-sized particles (Weber et al., 1995; Kulmala et al., 2004; Guo et al., 2012). For example, under the condition of photolysis, the chemical reaction of SO_2 with O_3 in the atmosphere would be as follows: $3\text{SO}_2 + 3\text{H}_2\text{O} + \text{O}_3 = 3\text{H}_2\text{SO}_4$ (4) (Hao et al., 2015). Thus, the higher concentrations of O_3 and SO_2 observed in NPF events indicated that O_3 and SO_2 might be main potentially precursor gases for our NPF events, but more researches are needed to confirm these findings.

4. Summary and conclusions

An intensive field measurement of aerosol particles and related quantities (including trace gases and meteorological data) were measured at the top of Mt. Huang (1840 m a.s.l), southeast China in 2008. The measurements covered the size range from 10 nm to 10 μm . The results obtained in this study could provide new insights into the evolution of particle number size distribution and new particle formation events for regional and global climate models.

The average particle concentration was $2.35 \times 10^3 \text{ cm}^{-3}$, which was lower than the values measured at other rural and mountainous regions of the world. On average, the ultrafine particles ($<0.1 \mu\text{m}$) represented 70.5% of the total particle number concentration. The overall number size distribution showed obvious diurnal variation with bimodal and single modal during the day and night time respectively. Excluding the accumulation mode particles, the average daytime particle number concentrations were prominently higher than those measured at nighttime, suggesting

there was a diurnal pattern of changes between PBL and FT air at the top of Mt. Huang. The aerosol spectra could be classified into two categories. The first category (FCS) exhibited a clear diurnal cycle, with relatively higher particle number concentration ($3.19 \times 10^3 \text{ cm}^{-3}$) and smaller size (ultrafine particles accounting for 75.4%). The second category (SCS) presented less obvious diurnal cycle, with lower particle number concentration ($1.88 \times 10^3 \text{ cm}^{-3}$) and larger size (ultrafine particles account for 60.2%). Air mass sources, weather conditions, and NPF events were responsible for the differences of particle spectra. The highest (FCS) and lowest (SCS) Aiken mode particle number concentration were observed in air masses from the continent and coastal regions, respectively. On sunny days (FCS), temperature and RH were the main affecting factors for the aerosol spectrum change, while the main factor affecting on cloudy days (SCS) was the wind speed.

New particle formation events occurred quite frequently at Mt. Huang, amounting for 37.5% of all the effective measurement days, and all events occurred on sunny days. The estimated formation rate J_{10} varied from 0.09 to $0.30 \text{ cm}^{-3} \text{ s}^{-1}$ with an average value of $0.23 \text{ cm}^{-3} \text{ s}^{-1}$, and the growth rate ranged from 1.42 to 4.53 nm h^{-1} with an average value of 2.90 nm h^{-1} . The events usually began at 10:00-11:00 LT characterized by the occurrence of a nucleation mode with a peak diameter of 10-20 nm. Wind speed, SO_2 and O_3 concentrations were all on average higher on NPF events days than on non-NPF event days. Temperature, RH, NO_x and CO concentrations were lower on NPF event days than on non-NPF event days. SO_2 and O_3 might be main potentially precursor gases for our NPF events. Southwest winds were more closely during the NPF events while south/southeast winds dominated on the non-NPF event days, in accordance with the back trajectory results. Our findings indicated that the NPF events were occurred more frequent and lasted longer at high altitude site, by comparing with the studies observed at the foot of Mt. Huang (Wang et al., 2014; Hao et al., 2015). The PBL development and the strong upslope wind occurred during daytime, the lower temperatures and pre-existing particle concentration at higher altitudes might be responsible to that phenomenon. However, these factors were not verified in this work since no simultaneous measurements were available at the top and foot of Mt. Huang. Thus long-term simultaneous measurements at different altitudes are needed in this region, especially to quantify the contributions of the PBL development and the upslope wind to NPF events. Future studies should also allow to confirm the main potential compounds participating in the nucleation and growth of particles at Mt. Huang.

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Fig.1. The location of Mt. Huang

Fig.2. Time series of particle number size distribution, total number concentration, temperature, relative humidity, wind speed and direction, air pressure, and precipitation intensity measured during study period. The shaded color curves are particle size distribution. Vertical bars are standard errors.

Fig.3. The average diurnal variation of number concentration of (a) total particle, (b) accumulation mode particle, (c) Aitken mode particle, and (d) nucleation mode particle. Vertical bars are standard errors.

Fig.4. The average particle number size distribution of daytime (solid) and nighttime (hollow).

Vertical bars are standard errors.

Fig.5. The average particle number size distribution of FCS (triangle) and SCS (circle). Vertical bars are standard errors.

Fig.6. The average particle number size distribution of FCS (triangle) and SCS (circle) during the daytime (solid) and nighttime (hollow) respectively. Vertical bars are standard errors.

Fig.7. 48-h air mass back trajectories arriving at Bright Summit for FCS, with 1400 m and 800 m arrival height respectively.

Fig.8. 48-h air mass back trajectories arriving at Bright Summit for SCS, with 1400 m and 800 m arrival height respectively.

Fig.9. Time series of the particle GMD with relative humidity and temperature in FCS.

Fig.10. Time series of the particle GMD with relative humidity and temperature in SCS.

Fig.11. Time series of wind speed with (a) the GMD and (b) the nucleation mode particles in SCS.

Fig.12. Contour plots of the average diurnal variations of particle number size distribution, the meteorological parameters and the concentrations of gaseous pollutants on 6 May 2008 at Mt. Huang. From top to bottom, the parameters are: (a) Particle size distribution; (b) Number concentrations of nucleation mode, Aitken mode and accumulation mode particles; (c) Temperature and RH; (d) Wind direction and speed; (e) NO_x and SO_2 ; (f) O_3 and CO . Vertical bars are standard errors.

Table 1 Introductions of sampling instruments

Instrument	Sampling type	Calibration	Reference
WPS	DMA	NIST SRM 1691 and SRM 1963- PSL sphere sizes ($0.269 \pm 0.007 \mu\text{m}$ and $0.1007 \pm 0.0020 \mu\text{m}$)	Liu et al. (2010)
	CPC		
	LPS	NIST-traceable PSL sphere sizes ($0.701 \mu\text{m}$, $1.36 \mu\text{m}$, $1.6 \mu\text{m}$, and $4.0 \mu\text{m}$)	
TEI Inc.	49i	zero air supplier (Model 111) O_3 calibrator (TE 49C PS)	Wang et al. (2013)
	48i	dynamic gas calibrator (Model 146)	
	43i	zero air supplier (Model 111)	
	42i	standard reference gas mixture (NO/CO in N_2)	

NIST, SRM, and PSL are the abbreviations of National Institute of Standards and Technology, Standard Reference Materials and Polystyrene Latex, respectively.

Table 2 Number concentration of aerosol particles in different size ranges measured at the top of Mt. Huang in southeast China from 22 April to 7 May, 2008.

	Size ranges (μm)	Mean $\pm \sigma$ (cm^{-3})	Max (cm^{-3})	Min (cm^{-3})	Ratio (%)
FCS (Samples: 1324)	0.01-0.1	2403 $\pm$ 1727	11763	284	75.4
	0.1-0.5	763 $\pm$ 392	2835	125	24.0
	0.5-2.5	20 $\pm$ 18	113	2	0.6
	2.5-10	0 $\pm$ 0	10	0	0
	0.01-10	3187 $\pm$ 1878	14623	494	
SCS (Samples: 1054)	0.01-0.1	1133 $\pm$ 1122	8687	200	60.2
	0.1-0.5	698 $\pm$ 320	2485	10	37.0
	0.5-2.5	52 $\pm$ 27	125	1	2.8
	2.5-10	0 $\pm$ 0	11	0	0
	0.01-10	1884 $\pm$ 1240	9769	253	
Total (Samples: 2849)	0.01-0.1	1657 $\pm$ 1541	11763	130	70.5
	0.1-0.5	664 $\pm$ 379	2835	10	28.2
	0.5-2.5	30 $\pm$ 27	125	1	1.3
	2.5-10	0 $\pm$ 0	11	0	0
	0.01-10	2351 $\pm$ 1714	14623	196	

FCS refers to the first category of aerosol continuous spectrum.

SCS refers to the second category of aerosol continuous spectrum.

Table 3 Comparison with the number concentration results in other mountainous sites.

Location	Size range (nm)	Number conc. (cm ⁻³)	Source
Himalayas, India	10-800	3420	Komppula et al. (2009)
Mt. Izaña, North Atlantic	13-600	1498	Rodriguez et al. (2009)
Mt. Norikura, Japan	9-300	300-1300	Nishita et al. (2008)
Mt. Jungfrauoch, Swiss alps	10-750	900	Weingartner et al. (1999)
Mt. TMS, Hong Kong, China	5.5-300	9500 in NPF days	Guo et al. (2012)
		5000 in non-NPF days	
Mt. Tai, China	10-100	5193	Gao et al. (2009)
	100-500	1419	
Mt. Waliguan, China	12-95	2640	Kivekäs et al. (2009)
	95-570	700	
Mt. Huang, China	10-100	1657	This work
	100-500	664	

Table 4 The ratios of daytime (09:00-20:00) to nighttime (21:00-08:00) average number concentrations of particles of each mode.

	nucleation	Aitken	accumulation	total
Daytime average/Nighttime average	10.80	1.49	1.06	1.51
Daytime median/Nighttime median	11.36	1.69	1.04	1.65

Table 5 Case statistics of aerosol particle formation and growth during observation.

Date	4/24	4/25	4/26	4/27	5/5	5/6	Average
Initial time	10:21	10:55	10:02	10:22	13:00	10:24	10:51±66 min
Duration	11:47	6:13	11:05	9:13	8:53	12:30	9:57±139 min
End time	22:08	17:08	21:07	19:35	20:53	22:54	20:38±123 min
Final diameter (nm)	35	38	42	30	53	74	45±16
J_{10} (cm ⁻³ s ⁻¹)	0.15	0.30	0.26	0.09	0.28	0.29	0.23±0.09
GR (nm h ⁻¹)	1.56	3.44	2.22	1.42	4.22	4.53	2.90±1.35
Air mass origin	SW	W	WSW	WNW	SE	SSW	SW

Table 6 Statistical summary of trace gases, meteorological parameters on NPF days and non-NPF days, listed is the average $\pm$ standard deviation of each quantity.

Parameter		NPF	Non-NPF
Trace gases (ppb)	O ₃	70.9 $\pm$ 6.5	60.8 $\pm$ 13.4
	SO ₂	5.6 $\pm$ 3.6	3.8 $\pm$ 3.0
	NO _x	1.8 $\pm$ 1.0	2.9 $\pm$ 1.4
	CO	261.5 $\pm$ 217.1	539.4 $\pm$ 184.0
Meteorological parameters	RH (%)	45.6 $\pm$ 10.4	83.9 $\pm$ 18.6
	T (°C)	10.9 $\pm$ 3.4	13.5 $\pm$ 2.4
	WS (m/s)	5.1 $\pm$ 1.7	5.0 $\pm$ 3.8
	WD	218.1 $\pm$ 72.6	196.5 $\pm$ 82.6

Date	4/24	4/25	4/26	4/27	5/5	5/6	Average
Initial time	10:21	10:55	10:02	10:22	13:00	10:24	10:51±66 min
Duration	11:47	6:13	11:05	9:13	8:53	12:30	9:57±139 min
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GR (nm h ⁻¹)	1.56	3.44	2.22	1.42	4.22	4.53	2.90±1.35
Air mass origin	SW	W	WSW	WNW	SE	SSW	SW

SO₂, O₃, low T and low RH favored the NPF