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MEASUREMENTS OF PARTICLE NUMBER SIZE DISTRIBUTIONS AND NEW PARTICLE FORMATION EVENTS DURING WINTER IN THE PEARL RIVER DELTA REGION, CHINA

TAN Hao-bo (谭浩波)[1, 2], YIN Yan (银 燕)[2], LI Fei (李 菲)[1], LIU Xian-tong (刘显通)[1], CHAN Pak-wai (陈柏纬)[3], DENG Tao (邓 涛)[1], DENG Xue-jiao (邓雪娇)[1], WAN Qi-lin (万齐林)[1], WU Dui (吴 兑)[1]

(1. Institute of Tropical and Marine Meteorology/Guangdong Provincial Key Laboratory of Regional Numerical Weather Prediction, CMA, Guangzhou 510080 China; 2. Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters, Nanjing University of Information Science and Technology, Nanjing 210044 China; 3. Hong Kong Observatory, Hong Kong China)

Abstract: Particle number size distribution (PNSD) between 10 nm and 20 μm were measured in the Pearl River Delta (PRD) region in winter 2011. The average particle number concentration of the nucleation mode (10–20 nm), Aitken mode (20–100 nm), accumulation mode (100 nm–1 μm) and coarse mode (1–20 μm) particles were 1 552, 7 470, 4 012, and 19 cm^{-3}, respectively. The volume concentration of accumulation mode particles with peak at 300 nm accounted for over 70% of the total volume concentration. Diurnal variations and dependencies on meteorological parameters of PNSD were investigated. The diurnal variation of nucleation mode particles was mainly influenced by new particle formation events, while the diurnal variation of Aitken mode particles correlated to the traffic emission and the growth process of nucleation mode particles. When the PRD region was controlled by a cold high pressure, conditions of low relative humidity, high wind speed and strong radiation are favorable for the occurrence of new particle formation (NPF) events. The frequency of occurrence of NPF events was 21.3% during the whole measurement period. Parameters describing NPF events, including growth rate (GR) and source rate of condensable vapor (Q), were slightly larger than those in previous literature. This suggests that intense photochemical and biological activities may be the source of condensable vapor for particle growth, even during winter in the PRD.

Key words: aerosol particle number size distribution; new particle formation; Pearl River Delta

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1 INTRODUCTION

Atmospheric aerosols directly affect the radiation balance of Earth's atmosphere by scattering and absorbing radiation, and also act as cloud condensation nuclei (CCN) which alter cloud droplet number concentration and affect the optical properties and lifetime of clouds, thereby indirectly affecting Earth's radiation balance (Chylek and Coakley [1]). Numerous pathological studies have shown that atmospheric aerosols are closely related to prevalence rates, hospitalization rates and even mortality rates for some diseases (Pope[2]; Atkinson et al.[3]), and ultrafine particles (UFP, aerodynamic diameter<100 nm) especially show greater impacts on health (Kreyling et al.[4]). Particle size distribution has more direct effects on health than mass concentration (Penttinen et al.[5]). Studies have shown that the size of aerosols is more relevant than their chemical composition in determining their ability to evolve into CCNs (Dusek et al.[6]). The fact that particle concentrations in accumulation mode show a high extinction efficiency can largely explain the fact that aerosols worsen visibility during haze (See et al.[7]). Standard particle monitoring usually focuses on mass concentration, however, particles with a diameter of less than 100 nm in urban areas generally account for a small proportion of overall mass but a vast majority of concentration. Therefore, it is more meaningful to perform observations on the particle size distribution (Seinfeld and Pandis[8]).

Hussein et al.[9] categorized aerosol number size distribution into four modes based on aerosol diameter measurements: nucleation mode (3-25 nm), Aitken mode (25-100 nm), accumulation mode (100-1 000 nm) and coarse mode (>1 μm). Different modes correspond to different optical characteristics, toxicological properties, sources, chemical composition, chemical evolution and removal processes (Whitby [10]). Currently, many

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Biography: TAN Hao-bo, Associate Professor, mainly undertaking research on atmospheric physics and environment.

Corresponding author: TAN Hao-bo, e-mail: hbtan@grmc.gov.cn

measurements on particle number size distribution (PNSD) have been started, covering various atmospheric environments around the world, including urban boundary layers (Birmili et al.[11]), rural areas (Dal Maso et al. [12]; Shen et al.[13]), background regions (Boulon et al.[14]; Weber et al.[15]), polar atmosphere (Laakso et al.[16]), ocean atmosphere (O'Dowd et al. [17]), and free troposphere (Venzac et al.[18]). Observations of PNSD in Niemi et al. [19] and Birmili et al. [20] have been ongoing for more than 10 years. Number concentrations of atmospheric aerosols, as a whole and for each mode, are affected by weather conditions, pollution emissions and movements of air masses, and display seasonal and diurnal variations (Laakso et al.[16]; Tunved et al.[21]). Particle number concentrations in urban atmospheres, especially those for Aitken mode particles, have diurnal variation mainly due to traffic emissions. Hence, traffic emissions were considered to be one of the most important sources of particles in the urban atmosphere (Wehner and Wiedensohler [22]; Hussein [23]). In addition, new particle formation (NPF) events are also an important source of particles (Stanier et al.[24]).

NPF events are referred to the dramatic increases in the number concentration of nucleation mode particles and their continued growth thereafter. During an NPF event, supersaturated vapor (e.g. sulfuric acid vapor) forms molecular clusters, which constitute the initial formation stage for particles, which then develop into measurable particles through various processes such as condensation and coagulation (Birmili and Wiedensohler[25]). Both sulfuric acid and organics can contribute to the subsequent condensational growth (Kulmala[26]). Currently, identification of NPF events is performed mainly based on number size distribution, so most of the studies concerning NPF events are based on variations in 3–25 nm particles (Kulmala et al.[27]). NPF can occur under both clean and polluted atmospheric conditions, though different locations produce significant variation in nucleation and growth processes due to variation in precursors, formation mechanisms and meteorological conditions (Stanier et al. [24]; Birmili and Wiedensohler [25]; Wu et al.[28]; Yue et al.[29]; Kulmala et al.[30]).

The Pearl River Delta (PRD) is one of China's three main economic zones. With the rapid growth of the urban population in Guangzhou in recent years along with accelerating industrialization and urbanization in this region, air quality in Guangzhou and the PRD region has been subject to widespread scrutiny, especially with regard to secondary pollutants such as fine particles and ozone pollution. As a result, problems caused by deterioration in air quality and visibility have become quite prominent and the PRD region is one of China's four top regions for severe haze (Chan and Yao[31]; Andreae et al.[32]; Zhang et al.[33]; Tan et al.[34]). Winter is the most haze-prone season in the PRD region (Tan et al.[34]; Wu et al.[35]; Chen et al.[36]). Although large-scale aerosol observations have been performed in the PRD region in the past [33], observations which attempt to link the characteristics of aerosol number size distribution in the winter and NPF events remain lacking. This paper presents an analysis of particle number size distributions during the winter in Guangzhou. Diurnal variations and dependencies on meteorological parameters of PNSD were investigated. This paper also analyzed the incidence of NPF events as well as favorable conditions and parameters describing NPF events during the measurement period.

2 MEASUREMENT SITE AND INSTRUMENTATION

Field experiments were conducted at the China Atmosphere Watch Network (CAWNET) station in Panyu, Guangzhou from November 10 to December 30, 2011. The Panyu station is located at the center of the PRD region and at the top of Dazhengang Hill (23.05° N, 113.34° E) with an altitude of about 150 m. Thus, the station represents the typical atmospheric conditions in this region. This site is surrounded by residential neighborhoods with no significant pollution sources nearby.

Atmospheric particle number size distribution (PNSD) was measured using both a scanning mobility particle sizer (SMPS 3936, TSI Inc.) and aerodynamic particle sizer (APS 3321, TSI Inc.). The SMPS was composed of a neutralizer (Kr–85), differential mobility analyzer (DMA 3081) and a condensation particle counter (CPC 3772) and this instrument was responsible for measuring the number size distribution of particles ranging from 10–500 nm in (Stokes) diameter. In an SMPS, sheath flow is performed in a closed loop mode, the ratio of sheath flow and sample stream flow is 6.5:1, and size-dependent pipe diffusion loss is corrected using the empirical formula defined in Willeke and Baron[37]. APS-measured aerodynamic diameter was then converted to Stokes diameter by assuming an aerosol particle density of 1.7 μg/m^3. A $PM_{2.5}$ impactor was installed on the outdoor air intake. Prior to entry into the APS and SMPS, sample aerosols were passed through a Nafion tube (Perma Pure Inc.) and dried to a relative humidity of less than 20%. There were 75 and 50 scanning channels in the SMPS and APS, respectively, and the time required for a single PNSD scan was 10 minutes. Since different sizes of particles have different sources, atmospheric evolution characteristics and life cycles, this paper categorized aerosols into four modes according to particle sizes [27]: Nucleation mode particles (Nuc): 10-20 nm, Aitken mode particles (Ait): 20-100 nm; Accumulation mode particles (Acc): 0.1-1 μm, and Coarse mode particles (Coa): 1–10 μm. Additionally, the prefix "N" and "V" denote number and volume concentrations of particles within the above size ranges, respectively.

In addition, meteorological parameters such as wind speed, wind direction, air temperature, relative humidity and ultraviolet radiation were also used in this paper.

3 RESULTS

3.1 *Particle number size concentration*

Figure 1 (a & b) shows the evolution of the particle number concentration during the measurement period. The total and different mode concentrations are shown. The average total concentration was larger during November (15 065 cm^{-3}) than during December (12 458 cm^{-3}), which was related to the lower average wind speed (1.7 m/s) in November than that in December (2.3 m/s) and more frequent occurrence of northerly wind in December (Fig.1c). From November to December, South China is usually affected by cold air masses moving southward. When cold air passes through the area, surface wind speed increases significantly, northerly wind component strengthens, relative humidity decreases and precipitation may occur (Fig.1 (c-g)). This can also promote the diffusion and clearance of pollutants. Afterwards, with the weakened high pressure system controlling South China, the weather becomes more stable and wind speed becomes weaker before a new round of cold air moves in [36]. Wu et al.[35] found that heavy pollution usually results from stagnant air masses during the aforementioned latter period over PRD. When the total concentration and relative humidity (RH) were relative high, the visibility was low, which is related to aerosol particle hygroscopic effect and heavy PM loading. The N_{tot} ranges from 2 458 cm^{-3} to 38 419 cm^{-3} with an average of 13 474 cm^{-3} during the measurement period. This number concentration is comparable to the concentration in Pittsburgh[24] and Atlanta (Woo et al.[38]), but lower than that in Beijing (Wu et al.[39]). It was also lower than that observed in this area around 2006 (Liu et al.[40]), which was consistent with the trend of PM reported by the Guangdong Atmospheric Component Bulletin.

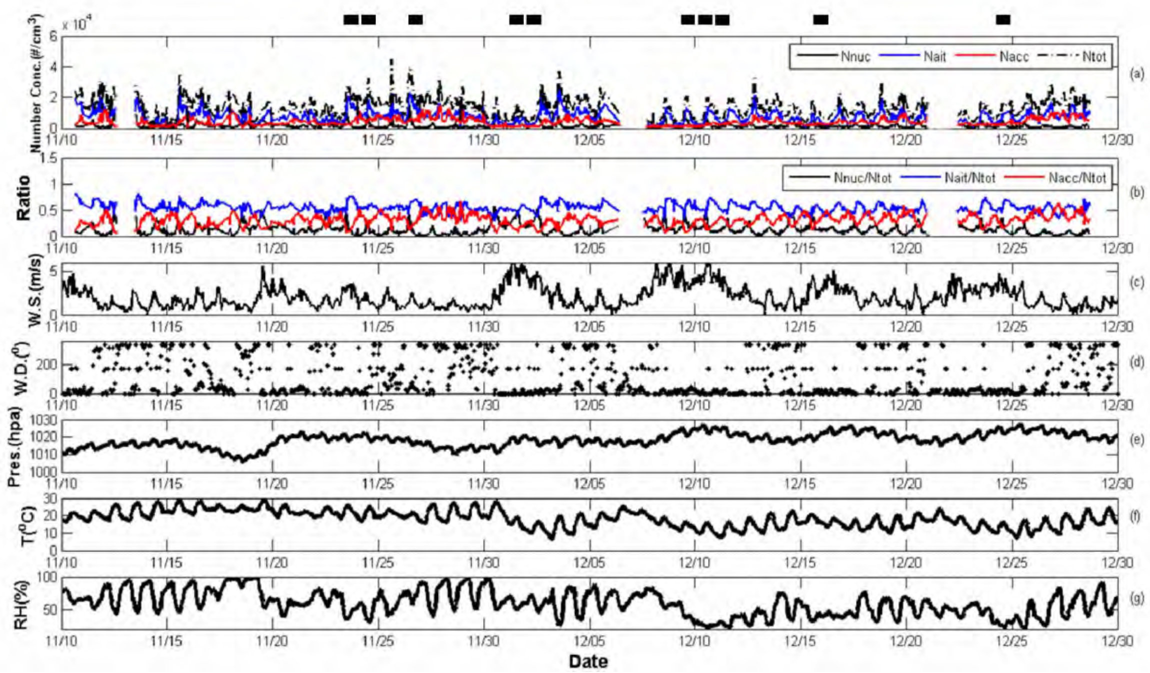

Figure 1. Time series of (a) particle number concentration, (b) ratio of number concentration of different modes to total concentration, (c)–(f) and main meteorological parameters measured at the measurement site during the measurement period. Black squares denote for the NPF events.

Figure 2 shows the mean and one standard deviation of particle number (upper panel), surface area (middle panel) and volume (bottom panel) size distributions during the measurement period. In this study, particle surface area and volume size distributions were calculated from the measured PNSD with an assumption of spherical particles. During the measurement period, PNSD showed distinct multi-modal patterns, with a majority of particles in Aitken mode and accumulation mode. N_{nuc}, N_{ait}, N_{acc} and N_{coa} accounted for 12.5%, 53.9%, 30.4% and 3.2%, respectively.

The peaks of Aitken mode were quite similar in November and December, which were at about 40 nm. The reason was that the Aitken mode particles are mainly associated with traffic activities in the urban area. The number concentrations for the Aitken mode and accumulation mode particles were higher in November than in December, and this can be explained by the meteorological conditions. As is aforementioned, the more frequent cold air coming from north resulted in good dispersion and deposition conditions of pollutants in December. In addition, the air mass back trajec-

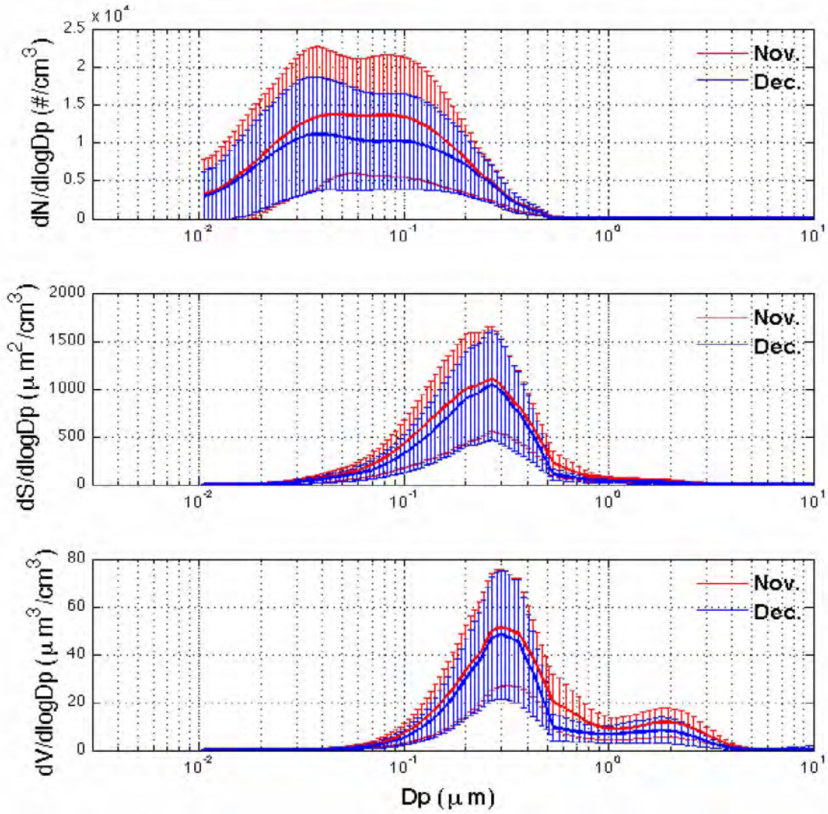

Figure 2. Particle number size distribution, surface area size distribution and volume size distribution in urban area of Guangzhou. The upper line and lower line of shaded area represent one standard deviation.

tories showed that air parcels spent more time in November over the PRD region.

The particle volume size distribution exhibited a distinct bimodal distribution, with a majority of the particle volume in the accumulation mode. The average V_{tot} was 29.2 $\mu m^3/cm^3$. The larger peak at about 300 nm accounted for over 70% of $PM_{2.5}$, by assuming homogenous particle density. A smaller peak appeared at 2 μm, which was more affected by construction, road dusts and biological sources. This illustrated that the accumulation mode particles were the main contributor to the PM pollution in the PRD region. Comparing to a previous study in 2006, the proportion of V_{coa} decreased. It indicated that the coarse mode particles from primary emission were decreasing, since the control of dust emissions from construction sites and roads has been effective in recent years.

3.2 *Diurnal variation of PNSD*

The averaged diurnal pattern of N_{tot} did not show big differences between November and December, and there were three peaks (Fig.3). Nucleation mode number concentration began to rise in the morning and reached a peak at around noon, thereafter declining in the afternoon. This diurnal variation was mainly caused by atmospheric NPF processes (gas-to-particle conversion). The occurrence of NPF events was closely related to meteorological conditions, and events tended to occur under low RH and sunny conditions [28]. Furthermore, NPF events were also correlated with pollutant concentration in the atmosphere. Since the PRD region usually has calm weather in the fall and winter, pollutants tend to accumulate and excessive coagulation of particles does not facilitate the formation and growth of particulate matter (see section 3.4). The diurnal variation of Nait showed a three-peak pattern. The morning peak occurred around 08:00 and resulted from rush hour traffic combined with relatively low mixed layer height. This morning peak was smaller than that in Beijing [39]. Possible explanation is that the starting time for mixed layer growth in Guangzhou is earlier than that in Beijing due to Guangzhou's earlier sunrise during winter. After 08:00, as human activity gradually reduced, the growth of particle concentration slowed down slightly. From 09:00 onward, because the concentration of the nucleation mode particles was continuously increasing, the nucleation mode particles were able to quickly grow to the Aitken mode through aging processes, including coagulation, collision and condensation, resulting in a rising Aitken mode particle concentration that reached a peak of 9 800 cm^{-3} at around 14:00. The third Aitken mode particle concentration peak occurred at 19:00–20:00 with a peak concentration of approximately 10 500 cm^{-3}. This peak was primarily related to nighttime human activities such as vehicle emissions and kitchen emissions from households and businesses. At the same time, there usually was a warm near-ground inversion layer during the nighttime, which resulted in a lower atmospheric boundary layer height that was not conducive to the diffusion of pollutants. The accumulation mode particles showed the longest average lifespan in the atmosphere and their concentration was also the most stable. Accumulation mode particles are mainly affected by changes in the mixed layer and their concentration was low during the day and high during the night. Nacc was lower than N_{ait}, and this was due to intense traffic emissions in Guangzhou.

The diurnal evolution of PNSD (Fig.3) showed an obvious growth process starting at 10:00, which was probably caused by NPF. The nucleated particles grew to larger sizes due to the aging process within several hours, which resulted in the increase of Aitken mode and subsequent accumulation mode particles.

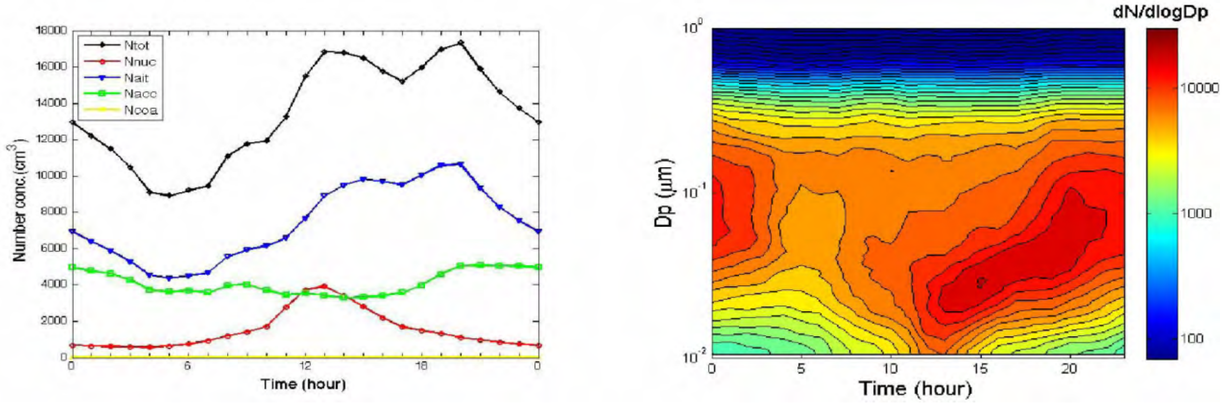

Figure 3. Average diurnal variation of different mode particles (left panel) and PNSD (right panel) during the measurement period.

3.3 *Dependence of PNSD on surface meteorological parameters*

The dependency of size-fractionated particle number concentration on wind speed, RH and temperature was investigated (Fig.4). It was determined that N_{ait}, N_{acc} and N_{tot} decreased as wind speed increased while Nnuc increased as wind speed decreased. When wind speed exceeded 4 m/s, number concentration for each mode was maintained at a stable level. Pollution was usually accompanied by low wind speed in which N_{ait} and N_{acc} concentrations were high. Strong coagulation scavenging produced by high concentration of accumulation mode leads to low number concentration of the nucleation mode particles. Furthermore, nucleation process is depressed by condensable vapor condensing rapidly among larger Aitken mode and accumulation mode particles (Mönkkönen et al.[41]). During passage of a cold front, wind speed suddenly increased along with a sharp decline in RH. At this time, dilution and diffusion effects from clean air lowered Nait and Nacc, thus weakening coagulation and condensable vapor via surface scavenging, which favored new particle formation. This result was similar to that of a previous study carried out in Beijing[39]. The reasons that N_{acc} increased as RH rose included: (1) when the area was shifting from colder high pressure to a ridge of weak high pressure or a uniform pressure field the average RH would gradually increase, thus the Aitken mode particles spent more time in the area and grew from the accumulation mode particles; (2) hygroscopic growth of aerosols increases the probability of forming the accumulation mode particles via collisions (Tan et al.[42]). Fig.2 shows that the accumulation mode particles are the main contributor of extinction coefficient and PM mass. Hence, low wind speed and high RH are two of the key factors leading to poor air quality and low visibility.

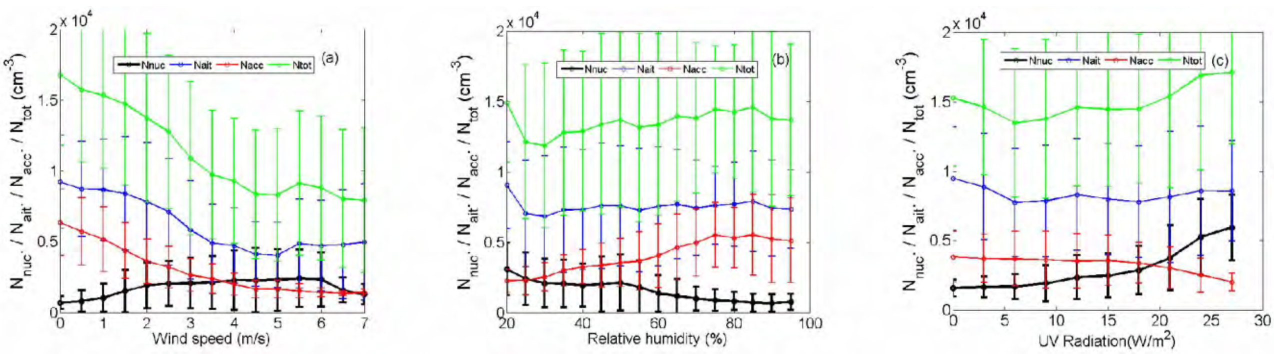

Figure 4. Dependence of size-fractionated particle number on (a) wind speed, (b) relative humidity and (c) UV radiation.

Figure 4c shows how the various modes change with the UV radiation intensity. N_{nuc} rises remarkably with the increase of UV radiation intensity, indicating that even in the winter season of Guangzhou, UV radiation causes more severe photochemical activities. This results in the generation of more condensable vapor such as sulfuric acid and low volatile organic compounds. N_{acc}, on the other hand, shows a decreasing trend after UV radiation has reached certain intensity. This is postulated to be due to the diurnal variation of the UV radiation intensity in relation to the boundary layer height. In general, the UV radiation intensity is the highest in the early afternoon when the boundary layer height has reached the highest value. When the N_{acc} amount is not changing too much, its concentration is decreasing with the lifting of the boundary layer height. Overall speaking, strong UV radiation facilitates the formation of new particles.

3.4 *New particle formation events*

The criteria used to discern NPF events were based on the burst of N_{nuc} [25], and an improved and clearer criteria was given by Dal Maso et al.[43]. Given the detection limit for the instruments used (10 nm), NPF criteria were defined as follows for the present study: (1) significant appearance of new modal particles from the nucleation mode (10–20 nm) in the particle number size distribution; (2) nucleation mode manifested in the atmosphere for more than 2 hours and in company of particle growth; (3) NPF events where particles grew to above 10 nm. Moreover, this study further divided these NPF events into two categories: Class I: events in which parameters (particle growth rate and particle formation rate) could be calculated with a high confidence level; Class II: events in which parameters were not calculable or fluctuated (Fig.5 shows an example on December 2).

During the measurement period of 2011, 10 NPF events (as listed in Table 1) out of 47 days were identified in Guangzhou. The frequency of occurrence (21.3%) was comparable to that in October 2004 at Xinken (25%[40]) and in July 2006 at Guangzhou (10%-25%, see Yue et al.[44]). Among the observation data, Class I events were selected for the purposes of calculating various parameters. Parameters describing NPF events including nucleation rate (J_{nuc}) of new particles (10–20 nm), growth rate (GR), condensational sink (CS) and source rate of condensable vapor (Q) were calculated with similar method as Dal Maso[43] and Wu et al.[28]. Table 2 lists the NPF parameters for this observation and those in some other literatures. The J_{nuc} values were in the range of 0.53–1.55 $cm^{-3}\ s^{-1}$, which were lower than those obtained at Xinken in October 2006 and at Beijing during summer. This may have been due to the fact that the instruments were unable to detect particles <10 nm in size, causing an underestimation of the number concentration of newly formed particles between 3–10 nm in size. GR was ranged from 3.5–8.5 nm/h and was comparable to those typical GR observed[30]. However, GR and Q were slightly higher than those observed in Beijing, indicating photochemical and biological activities in Guangzhou were more active than those in Beijing. GR depends on temperature and available condensable vapors, and it has been suggested that H_2SO_4 condensation typically accounts for only 10%–30% of the observed growth (Boy et al.[45]) compared to the VOCs which account for more than 70% of the materials for particle growth. The relative contribution of H_2SO_4 and VOCs in particle growth requires further study. CS is related to particle's surface area concentra-

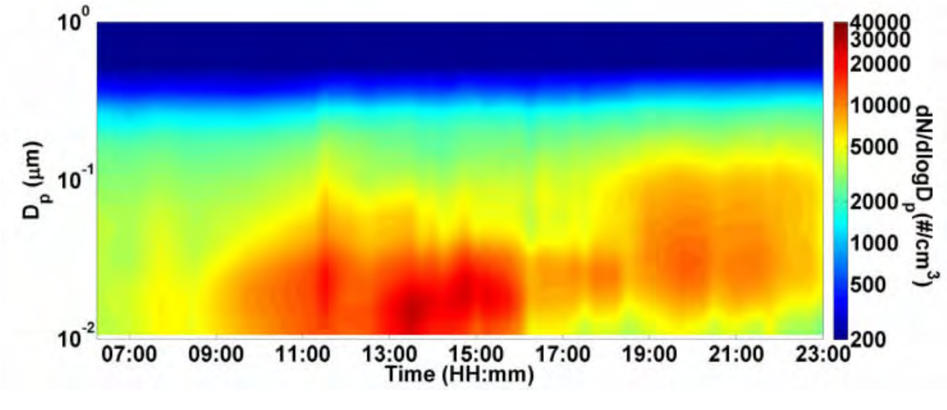

Figure 5. An example (December 2) of Class II NPF event.

Table 1. Dates of 10 NPF events.

Case	Time period (2011)	Type	Case	Time period (2011)	Type
1	1100 to 1400 Nov. 23	Class II	6	1000 to 1400, Dec. 9	Class I
2	1200 to 1400 Nov. 24	Class I	7	1200 to 1400, Dec. 10	Class I
3	1200 to 1300 Nov. 27	Class II	8	1200 to 1500, Dec. 11	Class I
4	1100 to 1400, Dec. 1	Class II	9	1200 to 1500, Dec. 16	Class II
5	1100 to 1400, Dec. 2	Class II	10	1200 to 1500, Dec. 24	Class I

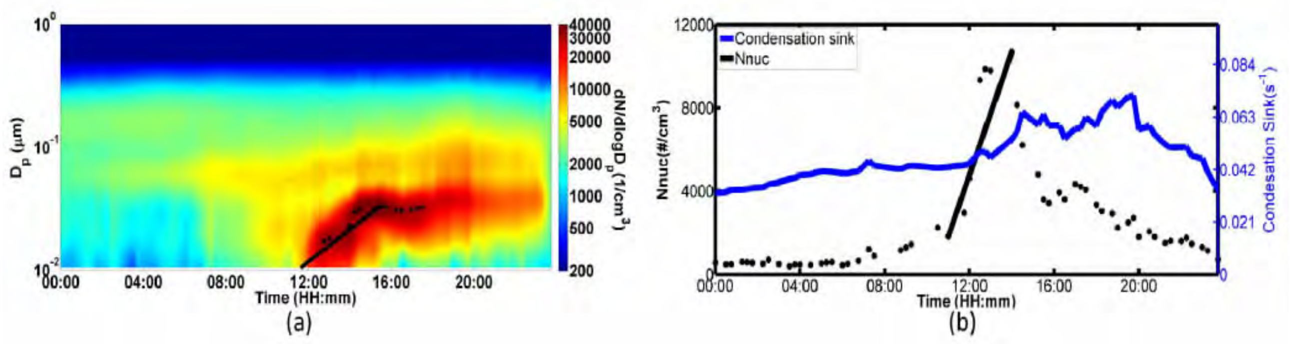

Figure 6. Parameters of an NPF event on December 9. (a) The black spot is the number mean diameter, while the solid black line denotes the growth rate during the event; (b) The solid black line denotes the nucleation rate of new particles during the event and the solid blue line denotes the condensational sink.

Table 2. Summary of Parameters Describing the NPF events.

Time & Location	Type	J_{nuc} (cm^{-3} s^{-1})	GR (nm h^{-1})	CS (10^{-2} s^{-1})	Q ($10^6 cm^{-3}$ s^{-1})
Nov. 24, 2011, Guangzhou	Urban	0.97	8.5	8.2	9.7
Dec. 9, 2011, Guangzhou	Urban	0.82	5.9	4.3	3.5
Dec. 10, 2011, Guangzhou	Urban	1.55	3.7	3.8	1.9
Dec. 11, 2011, Guangzhou	Urban	0.53	3.5	4.8	2.3
Dec. 24, 2011, Guangzhou	Urban	0.57	3.7	6.6	3.4
2004-2005, Beijinga	Urban	3.3–81.4	0.1–11.2	0.6–6.1	0.02–9.7
Oct. 26 to Nov. 9, 2002, New Delhi[b]	Urban	3.3–13.9	11.6–16.0	5–7	9–14
Jul. 1 to Jul. 19, 2002, Marseille[c]	Urban	–	1.1–8.1	0.32–1.5	0.087–1.3
Jun. 1995, England[d]	Coastal	0.91	1.3	–	–
Jan. 2000/2001, Antarctica[c]	Polar	–	0.3–2.7	0.02–0.96	0.0009–0.02
1997 to 2001, Hyytiala[c]	Rural	–	1.3–5	0.02–0.7	0.0005–0.69

Notes: a: Wu Z et al.[28], b: Mönkkönen P et al.[46], c: Kulmala M et al.[47], d: Harrison R M et al.[48].

tion. Before an NPF event, CS was maintained at a lower level and gradually increased when new particles formed and grew (Fig.6 shows an example on December 9).

4 CONCLUSIONS

PNSD between 10 nm and 20 μm was measured in the Pearl River Delta region in winter 2011. The average particle number concentration of nucleation mode (10–20 nm), Aitken mode (20–100 nm), accumulation mode (100 nm^{-1} μm) and coarse mode particles(1–20 μm) were 1 552, 7 470, 4 012 and 19 cm^{-3}, respectively. Particle number concentrations in PRD were noticeably lower than those in other China's mega cities and that observed in this area around 2006. Volume concentration of accumulation mode particles with peak at 300 nm accounted for over 70% of the total volume concentration.

Diurnal variation and dependency on meteorological parameters of PNSD were investigated. The diurnal variation of nucleation mode particles was mainly influenced by NPF events, while the diurnal variation of the Aitken mode particles correlated to traffic emissions and growth process of the nucleation mode particles. When the PRD region was controlled by cold high pressure, conditions of low relative humidity, high wind speed and strong radiation were favorable for the occurrence of NPF events.

The frequency of occurrence of NPF events was 21.3% during the whole measurement period. Parameters describing NPF events, including growth rate (GR), condensational sink (CS) and source rate of condensable vapor (Q), were slightly larger than those in previous literatures. This suggests intense photochemical and biological activities may be the source of condensable vapors for particle growth, even during winter in the PRD.

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