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Characteristics of regional new particle formation in urban and regional background environments in the North China Plain

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Abstract

Long-term measurements of particle number size distributions were carried out in the North China Plain both at an urban background site (Peking University, PKU) and a regional Global Atmospheric Watch station (Shangdianzi, SDZ) from March to November in 2008. In total, 52 new particle formation events were observed simultaneously at both sites, indicating that this is a regional phenomenon in the North China Plain. On average, the mean condensation sink value before the nucleation event start was 0.025 s^{-1} in the urban environment, which was 1.6 times higher than that at regional site. However, higher particle formation and growth rates were observed at PKU (10.8 $cm^{-3} s^{-1}$ and 5.2 $nm h^{-1}$) compared with those at SDZ (4.9 $cm^{-3} s^{-1}$ and 4.0 $nm h^{-1}$). These results implied that more precursors are needed to participate in the nucleation process to observe the occurrence of new particle formation event in a more polluted urban environment. Different from the observations in clean environments, the background condition of the observed nucleation events in the North China Plain could be characterized as the co-existing of the higher source and sink. The condensational growth of newly formed particles results in an increase in the particle mass concentration, particle light scattering coefficient, and CCN number concentration, with consequences on climate effects and air quality. In 34 investigated new particle formation cases at both sites, a significant particle nucleation and subsequent growth over a sufficient long time period were observed and investigated in terms of the particle light scattering and the number concentration of "potential" CCN. The results revealed that the new particle formation increases the particle light scattering coefficient and CCN number concentration in the North China Plain by factors in the range of 6.3–7.6 and 5.6–8.7, respectively. Moreover, the potential contribution of anthropogenic emissions to the CCN number concentration is more than 50 %, which should be drawn more attentions in the regional and global climate model, especially in the polluted urban areas.

1 Introduction

Impacts of aerosol particles on the earth-atmosphere system, including the climate, ecosystem, air quality and public health determine its highlight topics in atmospheric environmental research (IPCC, 2007). New particle formation (NPF), through the nucleation of gas phase species and continuous condensational growth, is an important source of aerosol particles in global scale (Yu et al., 2008; Merikanto et al., 2009). These newly formed particles can grow into sizes in which they can act as potential cloud condensation nuclei (CCN) and in this way influence the regional and global climate system indirectly (Lohmann and Feichter, 2005). On the other hand, in environments such as the North China Plain with a high potential of secondary aerosol formation, the particle light scattering coefficient can also be significantly increased as shown in Wiedensohler et al. (2009).

NPF events have been observed in a variety of atmospheric environments including the stratosphere (Lee et al., 2003), remote polar area (Park et al., 2004), high altitudes mountains (Weber et al., 1997; Venzac et al., 2008), inside boreal forest (Dal Maso et al., 2005), coastal environments (O'Dowd et al., 2002), continental rural and suburban regions (Birmili et al., 2003; Petäjä et al., 2007), as well as urban environments (Stanier et al., 2004; McMurry et al., 2005; Salma et al., 2011) and very polluted megacities such as Mexico City (Dunn et al., 2004), New Delhi (Mönkkönen et al., 2005) and Beijing (Wu et al., 2007). Meanwhile, evidences also revealed that ambient new particle formation might take place on a larger regional scale (Tunved et al., 2003; Dal Maso et al., 2007; Hussein et al., 2009; Bae et al., 2010; Jung et al., 2013). Kulmala et al. (2004) summarized the formation and growth properties based on over 100 field campaigns. The results showed that the typical particle growth rates in nucleation mode range from 1 to 20 $\mathrm{nm\,h^{-1}}$ in mid-latitudes; whereas particle formation rates vary widely depend on the air mass type of sampling site. However, most of these measurements were conducted in Europe and North America or they were limited for a short

time period, the long-term observations, especially carried out in developing countries, were rare.

In China, long-term continuous measurements of particle number size distributions have been first performed at Beijing urban (Peking University, PKU) since March 2004 (Wehner et al., 2004) and at regional background station Shangdianzi (SDZ) since March 2008 (Shen et al., 2011). High frequencies of NPF events were observed around Beijing region. In addition, the NPF events also had been observed at marine area (Lin et al., 2007), high-elevation mountains (Li et al., 2011; Guo et al., 2012), rural sites of Pearl River Delta (Liu et al., 2008; Wang et al., 2013c), suburban environments in Yangtze River Delta region (Gao et al., 2009; Herrmann et al., 2013) and Lanzhou (Gao et al., 2011), as well as urban Shanghai (Du et al., 2012). These observations were however independent and only based on for a short-term observations, no information on the long-term measurements of new particle formation events in a regional scale has been reported yet.

Regional NPF events have been observed in Beijing area based on the simultaneous long-term measurements of particle number size distributions both at PKU and SDZ sites from March to November 2008. In this study, we are aiming to investigate the relationship of NPF events between polluted urban and regional background environments. This is the first study to characterize regional NPF events in China. The characteristics are compared and the influences of NPF event on the aerosol direct (particle light scattering coefficient) and indirect (CCN number concentration) effects are estimated.

2 Measurements

2.1 Sampling site

The urban sampling site PKU is located in the northwest of Beijing center, outside the fourth-ring road. The observatory is set on the sixth roof of an academic building on

the campus of Peking University (39.99° N, 116.31° E; 50 m a.s.l.). Two major roads with heavy traffics at the east and south of the site are respectively 200 m and 500 m away. PKU site is assumed as representative of the Beijing urban environment. Detailed descriptions of the measurement site we refer the reader to Wu et al. (2008).

SDZ site (40.65° N, 117.28° E, 287 m a.s.l.) is one of the regional stations of the Global Atmosphere Watch (GAW) Program in China. This station is located in the north part of the North China Plain, about 12 km northeast of the PKU site. The sampling site is situated on the south slope of Yanshan Mountains, in the valley with a northeast-southwest orientation. There are few local anthropogenic pollution sources surrounding the SDZ, which could reflect the regional background situation for the North China Plain. Detailed information on the SDZ site can be found in Shen et al. (2011).

2.2 Instrumentation

Number size distributions of atmospheric particles in the size range 3–900 nm at PKU site and 3–850 nm at SDZ site were both measured by dual mobility particle size spectrometer TROPOS-type TDMPS (Twin Differential Mobility Particle Sizer) (Birmili et al., 1999). The whole system consists of two parallel Differential Mobility Analyzers (DMAs) and two Condensation Particle Counters (CPCs) (model 3010 and model 3025, TSI Inc., St. Paul, MN, USA). The relative humidity (RH) within the whole system was kept below 40 % both in the inlet line and in the sheath air cycle to obtain the number size distributions of dry particles. In addition, the particle number size distributions were corrected for particle losses inside the mobility particle size spectrometers and in the sampling configuration, following the method of the "equivalent length" as described in Wiedensohler et al. (2012).

2.3 Data analysis

2.3.1 Classification scheme

The event classification criterion is based on the method described by Dal Maso et al. (2005). However, the nucleation events are only divided into two groups in this study. The criterion for discerning an intensive nucleation event day (Class I) is the burst of new particles in the nucleation mode size range (3–25 nm) and the burst should last several hours with a clear growth pattern. On some days, the formation of new particles was observed without subsequent growth. These cases are more like the sudden burst of freshly nucleated particles, which are grouped into Class II. Besides these two kinds of NPF events, the sporadic occurrence of newly formed particles in the nucleation mode size range was also seen, however, the situation is not fulfill the criteria for the event days. These days are represented as "undefined".

2.3.2 Particle formation rate (FR) and growth rate (GR)

The lowest detecting particle size measured by the mobility particle size spectrometer is 3 nm. Hence, apparent formation rate (J_3) can be expressed as:

$$J_3 = \mathrm{d}N_{\mathrm{nuc}}/\mathrm{d}t + F_{\mathrm{coag}} + F_{\mathrm{growth}} \quad (1)$$

Here, N_{nuc} is the number concentration of nucleation mode particles. F_{coag} and F_{growth} represent the loss of particles due to coagulation and the flux of particles out of the specified size range, respectively. The second term in Eq. (1) can be calculated according to:

$$F_{\mathrm{coag}} = \mathrm{CoagS}_{\mathrm{nuc}} N_{\mathrm{nuc}} \quad (2)$$

in which $\mathrm{CoagS}_{\mathrm{nuc}}$ is the coagulation sink of nucleation mode particles. In this study, we consider the reference size for the coagulation sink to be the geometric mean diameter

of nucleation mode (8 nm). $\mathrm{CoagS}_{\mathrm{nuc}}$ is defined as:

$$\mathrm{CoagS}(D_\mathrm{p}) = \int K(D'_\mathrm{p}, D_\mathrm{p}) n(D'_\mathrm{p}) dD'_\mathrm{p} \quad (3)$$

Here, $K(D'_\mathrm{p}, D_\mathrm{p})$ is the coagulation coefficient of particles with diameters D_p and D'_p according to Fuchs (1964). The newly formed particles rarely grow beyond 25 nm before formation ended, and the last term can be neglected.

The observed particle growth rate is calculated based on the "log-normal distribution function method" described by Kulmala et al. (2012). Typical particle number size distributions are fitted by a least-square log-normal fitting method yielding parameters of 2–3 log-normal modes. Thus, the temporal variation of mean geometric diameters could be obtained and the particle growth rate can be estimated ($\mathrm{GR} = \mathrm{d}D_\mathrm{p}/\mathrm{d}t$, given in $\mathrm{nm\,h^{-1}}$).

Moreover, we should keep in mind that the determinations of start and end times of nucleation events in Class II type are difficult, which might cause huge uncertainty when calculating the particle formation and growth rates. Therefore, in this study we only focus on the nucleation event in Class I type, which shows the clear nucleation and growth diurnal patterns.

2.3.3 Condensation sink (CS), condensable vapor concentration (*C*) and source rate (*Q*)

The condensation sink, describing how rapidly vapor molecules can condense onto the particles, is used to represent the pre-existing particles concentration (Kulmala et al., 2001). Its value could be calculated using Eq. (4):

$$\mathrm{CS} = 2\pi D \sum \beta D_\mathrm{p} N \quad (4)$$

Here, D is the diffusion coefficient of the condensing vapor, β is the transitional regime correction factor, D_p is the particle diameter and N is the corresponding particle number concentration. However, it has to be noted that in this study, the CS values are

achieved based on the dry particle number size distributions, which could not necessarily represent ambient wet condition well, especially during the humid summertime. Hence, the presented CS values might be underestimated compared with real ones. The uncertainty caused by the typical effect of hygroscopic growth on CS is between 5 and 50 % as shown in previous research (Kulmala et al., 2001).

The condensable vapor concentration C and its source rate Q could be achieved based on the growth rate and the condensation sink (Kulmala et al., 2001). The growth rate depends on the amounts of condensable vapors in the atmosphere. Hence, the vapor concentration could be obtained by:

$$C = C_{\mathrm{GR=1\,nm\,h^{-1}}} \times \mathrm{GR} \quad (5)$$

Here, $C_{\mathrm{GR=1\,nm\,h^{-1}}}$ is the vapor concentration required for growth rate of $1\,\mathrm{nm\,h^{-1}}$, recent study (Nieminen et al., 2010) gave Eq. (6) to get its value:

$$C_{\mathrm{GR=1\,nm\,h^{-1}}} = \frac{2\rho_\mathrm{v} d_\mathrm{v}}{\gamma m_\mathrm{v} \Delta t} \cdot \sqrt{\frac{\pi m_\mathrm{v}}{8kT}} \cdot \left[\frac{2x_1 + 1}{x_1(x_1+1)} - \frac{2x_0+1}{x_0(x_0+1)} + 2\ln\left(\frac{x_1(x_0+1)}{x_0(x_1+1)}\right)\right] \quad (6)$$

Here we assume the properties of the condensable vapors are similar to gaseous sulfuric acid, which is proved to be the key component in nucleation process (Berndt et al., 2005; Sipila et al., 2010; Chen et al., 2012). The mass (m_v) and density (ρ_v) of hydrated sulfuric acid vapor molecule utilized in this study are 135 amu and $1650\,\mathrm{kg\,m^{-3}}$, respectively (Wang et al., 2013b).

With a pseudo steady state assumption, the vapor source rate can be estimated as

$$Q = \mathrm{CS} \cdot C \quad (7)$$

Where Q is the condensable vapor source rate in unit $\mathrm{cm^{-3}\,s^{-1}}$.

3 Results and discussions

3.1 Occurrence of new particle formation event

The investigation time period is from March to November 2008, during which the simultaneous measurements of particle number size distributions were performed at both sites. The numbers of days with valid data were 253 at PKU and 223 at SDZ, respectively, accounting for 92 % and 81 % to the total 275 investigated days (Table 1). The remaining days were either completely missing or have poor data quality. The frequencies of NPF event were 38 % and 39 % in classified days at urban and regional sites, respectively. These frequencies are similar with that observed in Beijing between March 2004 and February 2005 (Wu et al., 2007). One third of nucleation events at PKU were the Class II type, which was higher than that at SDZ (21 %). This might due to the influences of local emissions at urban site. In total, 52 NPF events were observed simultaneously at both places based on 207 valid days with simultaneous measurements, suggesting the new particle formation is a regional phenomenon over the North China Plain.

The similar "U-shape" seasonal variation patterns of NPF event occurrence were observed at both sites, as shown in Fig. 1. The number of events was highest in spring month (March–May), with 43 cases, occurring at both PKU and SDZ, respectively, accounting for 45 % and 49 % in total number of observed NPF events. More events were observed in spring may due to a higher frequency of strong wind from northern China, which favors to remove the pre-existing particles in the atmosphere and further lead to the occurrances of NPF event (Wu et al., 2008). During the summertime, high temperatures and RHs are not propitious for NPF events. In addition, slowly moving air masses from southern, which dominate in the summertime, always contains more accumulation mode particles, resulting in pollution episodes (Wehner et al., 2008; Wang et al., 2013a). Hence, few events (22 at PKU and 10 at SDZ) were observed during this time period. More NPF events were observed in autumn at SDZ (43 cases) compared with that at PKU (31 cases), especially in October and November. This is because that the

heavy pollution episodes always take place in the urban Beijing in this season. During the episode, the local meteorological condition (high temperature, moderate RH and calm wind) leads to very poor dispersion of pollutants and results in the local emissions are trapped in the urban atmosphere (Wu et al., 2008). On the contrary, the pre-existing particles are less at SDZ site, which favors to produce more events happening.

3.2 Nucleation event characterization

Individual and mean formation rates of aerosol particles with a diameter of 3 nm are shown in Fig. 2a. They varied between 2.2 $cm^{-3} s^{-1}$ and 34.5 $cm^{-3} s^{-1}$ at PKU as well as 0.4–24.5 $cm^{-3} s^{-1}$ at SDZ. In addition, the mean value was 10.8 $cm^{-3} s^{-1}$ at PKU, which was factor two higher than that at SDZ (4.9 $cm^{-3} s^{-1}$). The formation rates at both sites were significantly higher than the observations in clean environments such as 0.8 $cm^{-3} s^{-1}$ in Hyytiälä (Dal Maso et al., 2005) and 1 $cm^{-3} s^{-1}$ in Hohenpeissenberg (Birmili et al., 2003). However, the values at PKU are also much higher than observed at other urban sites such as Helsinki, 2.4 $cm^{-3} s^{-1}$ (Hussein et al., 2008) and Budapest, 4.2 $cm^{-3} s^{-1}$ (Salma et al., 2011), as well as in similar magnitude with study in New Delhi, 3.3–13.9 $cm^{-3} s^{-1}$ (Mönkkönen et al., 2005) and St. Louis, 17 ± 20 $cm^{-3} s^{-1}$ (Qian et al., 2007). The highest formation rates were shown at both sites in spring (Fig. 2b), which can be explained by the favoring meteorological conditions such as low temperature and humidity as well as strong north wind, bringing the clean air mass in this season. The growth rates ranged from 2.5 $nm\,h^{-1}$ to 15.3 $nm\,h^{-1}$ with a mean and standard deviation of 5.2 ± 2.2 $nm\,h^{-1}$ at PKU site, which was slightly higher than the values at SDZ (1.0–9.7 $nm\,h^{-1}$, 4.0 ± 1.7 $nm\,h^{-1}$). They essentially fit into the range of typical particle growth rate 1–20 $nm\,h^{-1}$ (Kulmala et al., 2004). In contrary to the particle formation rate, the highest growth rate is observed in summertime during which the enhancement of photochemical and biological activities together with the stagnant air masses preventing exchange with cleaner air contributed to the particle growth. This is consistent with a previous study (Wu et al., 2007).

More pre-existing aerosol particles (presented as CS in this study) were found at urban environment (see Fig. 3). The mean CS values on NPF event days were $0.027 \pm 0.021\,s^{-1}$ at PKU and $0.020 \pm 0.020\,s^{-1}$ at SDZ, respectively, which were much lower than those on none-event days ($0.047 \pm 0.024\,s^{-1}$ and $0.026 \pm 0.018\,s^{-1}$). During NPF event days, the CS values were 5–10 times higher than that in European cities such as Marseille ($0.003–0.015\,s^{-1}$), Athens ($0.006–0.013\,s^{-1}$) and Helsinki ($0.006\,s^{-1}$), while the highest value was comparable with polluted areas like New Delhi ($0.050–0.070\,s^{-1}$) (Kulmala et al., 2005; Hussein et al., 2008). It is remarkable that the mean CS value before the nucleation events start (08:00–11:00 LT) at PKU site was $0.025\,s^{-1}$, which was close to the observations at SDZ during none-event days ($0.024\,s^{-1}$). This result implied that more precursors are needed for particle nucleation process to observe the occurrence of new particle formation event at polluted urban environment.

Table 2 lists the statistical results of condensation sink, condensable vapor concentration and its source rate. The seasonal variations of vapor concentration and its source rate showed that the highest values during summer months. This result is consistent with the previous study by Wu et al. (2007). The vapor concentration at PKU ($9.3 \times 10^7\,cm^{-3}$) was 1.3 times higher than that at SDZ ($7.1 \times 10^7\,cm^{-3}$) during the whole measurement period. This is easy to understand that more vapors should be involved in the particle nucleation and growth processes to preventing the newly formed particles captured by higher pre-existing particles in a polluted urban environment. As a result, the source rate of condensable vapor was on average 1.8 times higher at Beijing urban compared to the SDZ site. Overall, unlike the observations in clean environments, the background condition of NPF events in the North China Plain could be characterized as the higher source (vapor concentration) and higher sink (pre-existing particles), even in the background regional station. Hence, the nucleation mechanisms in this background situation need to be concerned in further research.

3.3 Influences on the light scattering and CCN production

Aerosol particles formed by homogeneous nucleation can grow to larger sizes in which they could affect the global radiation budget by light scattering directly (Stier et al., 2007). Furthermore, most field observations (Kuwata et al., 2008; Kuang et al., 2009; Wiedensohler et al., 2009) pointed out that a large fraction of these particles may act as CCN in the atmosphere and influence the regional or global climate system indirectly (Lohmann and Feichter, 2005). Due to the lack of direct measurements of particle light scattering coefficients (σ_{sp}), its values were reconstructed based on the measured dry particle number size distributions utilizing a Mie model (Bohren and Huffman, 1998). The refractive index used in the code was 1.55–0.55i, which was derived from the ambient measurement at a rural site (Yufa) around Beijing (Cheng et al., 2009). Meanwhile, quantitative determination of CCN number concentration is not possible without direct measurements or detailed information on the volume fractions of chemical compositions. Since such data are not available during the investigated period, we have to rely on some surrogate measures for the CCN number concentration. The idea to estimate the production of potential CCN is based on the assumption that particles larger than a certain diameter could be activated to form CCN (Lihavainen et al., 2003; Laaksonen et al., 2005; Kerminen et al., 2012). Three quantities, CCN_{45}, CCN_{90} and CCN_{200}, namely the number concentrations of particles larger than 45 nm, 90 nm and 200 nm in dry diameter, respectively, are selected to represent the "potential" CCN number concentrations. This selection can be considered justified, because the field observations during the HaChi (Haze in China) summer campaign in the North China Plain (Wuqing site) showed the activation diameters at super saturations of 0.058 %, 0.085 %, 0.18 %, 0.36 % and 0.72 % are 200 nm, 170 nm, 90 nm, 70 nm and 45 nm, respectively (Deng et al., 2011). To investigate the influences of NPF event to the aerosol direct and indirect effects, here we first determined time t_1 and t_2 when these parameters (σ_{sp}, CCN_{45}, CCN_{90} and CCN_{200}) reached the minimum and maximum during the timeframe for the determination of particle growth rate, respectively. The increase is then denoted from

the differences between their minimum and maximum, which are calculated by averaging over $t_1 \pm 0.5$ h and $t_2 \pm 0.5$ h to decrease the uncertainties. Even these estimated values could not represent the real condition, we could see the influences of new particle formation on optical properties and potential CCN productions.

In addition, it also should be clarified that this method might produce several potential misunderstanding on estimating the contributions of NPF event to the particle light scattering coefficient and CCN number concentration in the polluted urban environment. This is because that the intensive primary emissions by anthropogenic activity at urban environment can emit both the precursors and aerosol particles into the ambient atmosphere. On one hand, these precursors could be involved in the nucleation process and further enhance the production of newly formed particles. On the other hand, the emitted particles are in a wide particle size range, which might directly influence the particle light scattering coefficient or act as potential CCN. Nevertheless, these effects could be neglected at a regional background site such as SDZ. Hence, in this section, we only pay attention to the regional NPF events observed concurrently at both sites (total 34 events in Class I type). Moreover, considering that the particle light scattering coefficient might be affected by the other parameters such as particle chemical compositions and mixing state, which are totally different between urban and regional sites, hence the differences in diverse environments will not be discussed further in this study. The potential contributions of regional NPF event to production of CCN are firstly calculated for the regional and urban sites, and the gap is estimated as mainly contributed by the anthropogenic emissions at polluted urban environment.

Figure 4 showed an example that simultaneous NPF events observed both at PKU and SDZ on 26 September 2008. New particles were nucleated around 09:00 LT in the morning followed by a significant growth to larger particles ($\sim$ 80 nm) indicated by the "banana-shaped" temporal development of the particle number size distribution (Fig. 4a and b). The particle number concentrations of the nucleation mode ($N_{3–25}$) and of all particles ($N_{3–900}$) were shown in Fig. 4c and d. At PKU site, the maximum of $N_{3–25}$ and $N_{3–900}$ were 52 300 cm^{-3} and 60 300 cm^{-3}, respectively. In contrary, the fraction

of $N_{3–25}$ in $N_{3–900}$ could up to 93 % at the beginning of the nucleation event and the peak values were shown at the same time at SDZ. The mean condensation sink before nucleation events start (08:00–11:00 LT) was 0.007 s^{-1} at PKU, which was almost one order of magnitude higher than that at SDZ (0.001 s^{-1}). However, the particle formation and growth rates were 9.8 $cm^{-3}\,s^{-1}$ and 4.5 $nm\,h^{-1}$ at PKU, as well as 4.3 $cm^{-3}\,s^{-1}$ and 3.7 $nm\,h^{-1}$ at SDZ, respectively, indicating more condensable vapor should contribute to the particle nucleation and growth processes in polluted urban environment.

The estimated particle scattering coefficients (550 nm) and mass concentrations are presented in Fig. 4e and f. The mass concentration was calculated using measured particle number size distribution with assumed density of 1.7 $g\,cm^{-3}$. Obviously, strong correlations (R^2 = 0.99) were observed between particle scattering coefficients and particle mass concentrations at both sites. At PKU site, the particle light scattering coefficients and mass concentrations showed a significant increase from 15:00 to 23:00 LT, with the average growth rates 13.9 $Mm^{-1}\,h^{-1}$ and 4.5 $\mu g\,m^{-3}\,h^{-1}$, respectively. On the contrary, these two parameters exhibited a continuing elevation until to midnight at SDZ. The growth rates were 7.8 $Mm^{-1}\,h^{-1}$ and 1.7 $\mu g\,m^{-3}\,h^{-1}$, respectively. The analysis above indicated that the subsequent growth of nucleated particles resulted in an increase in particle light scattering coefficient and mass concentration, through which affected the climate and air quality.

The variations of CCN number concentrations were displayed in Fig. 4g and h. CCN_{45} showed a simultaneous increase with the nucleation mode particles at both sites, however, the enhancements in CCN_{90} and CCN_{200} were several hours later. The lowest CCN_{45}, CCN_{90} and CCN_{200} were 2200 cm^{-3}, 700 cm^{-3} and 160 cm^{-3} during the event, respectively, and gradually raised to the maximum values of 15 600 cm^{-3}, 5600 cm^{-3} and 900 cm^{-3} for PKU at midnight. The enhancement factors were 7.0, 7.8 and 5.5, respectively. Contrary to this, these three quantities increased by factors of 24.3, 13.6 and 10.5 at SDZ. The higher CCN enhancement factors at SDZ might due to the lower pre-existing particles at the initial time. However, in absolute term, the increasing CCN_{45}, CCN_{90} and CCN_{200} were 4900 cm^{-3}, 2600 cm^{-3} and 370 cm^{-3}

higher at urban site compared with regional site, respectively, suggesting that the potential contributions of anthropogenic emissions to CCN number concentrations were 37 %, 54 % and 50 % in this case.

On average, the regional new particle formation resulted in the enhancement of particle light scattering coefficient with 200 ± 25 Mm^{-1} at PKU and 100 ± 90 Mm^{-1} at SDZ, respectively, with factors of 6.3 ± 3.8 and 7.6 ± 6.6. The higher increment at urban environment might be attributed to the anthropogenic emission and more complex atmosphere. Meanwhile, we observed CCN_{45}, CCN_{90} and CCN_{200} increase by factors of 5.6, 6.0 and 5.6 at PKU, respectively, and 8.7, 7.0 and 6.5 at SDZ (see Table 3). This result indicated very clearly that atmospheric new particle formation is an extremely important source of new "potential" CCN over the North China Plain. Moreover, the potential contribution by the anthropogenic emissions to CCN number concentration should be concerned at the urban site, with average fraction in the range of 59–63 %.

4 Conclusions

To investigate the characters of regional new particle formation events in the North China Plain, simultaneously measurements of particle number size distributions had been carried out at urban site PKU and regional site SDZ from March to November in 2008. Both station have been equipped with a dual mobility particle size spectrometer, type TROPOS-TDMPS.

In total, the frequencies of nucleation event were 38 % and 39 % at urban and regional background sites, respectively, with 52 regional NPF events at both places during the whole measurement period. The seasonal variations of NPF event occurrence were similar at both sites, with more events in spring and lowest number in the summertime.

Apparent particle formation rates were in the range of 2.2–34.5 cm^{-3} s^{-1} at PKU, which was higher than that at SDZ site (0.4–24.5 cm^{-3} s^{-1}). The mean growth rates were 5.2 nm h^{-1} and 4.0 nm h^{-1} at PKU and SDZ, respectively. Before the nucleation

event starts (08:00–11:00 LT), more pre-existing particles were observed at polluted urban environment than that at the regional site. The mean values of condensation sink were 0.025 s^{-1} at PKU and 0.016 s^{-1} at SDZ, respectively, which were both significantly higher than the measurements in European cities. Overall, the background condition of NPF event in the North China Plain could be characterized as the higher precursor vapor concentration and pre-existing particles, even in the regional background environment.

The condensational growth of nucleated particles results in an increase in the particle mass concentration, scattering coefficient and CCN production. The increase of the particle light scattering coefficient resulting from the new particle formation event is a factor in the range of 6.3–7.6 in the North China Plain. Three quantities, CCN_{45}, CCN_{90} and CCN_{200}, were selected to estimate the "potential" CCN number concentrations. On the average of 34 selected cases with significant growth pattern, CCN_{45}, CCN_{90} and CCN_{200} number concentrations are enhanced increase by factors of 5.6, 6.0 and 5.6 at PKU, respectively, and 8.7, 7.0 and 6.5 at SDZ. In addition, the anthropogenic emissions may contribute more than half (59–63 %) of the CCN productions at the polluted urban environment, which should be paid more attention in the regional and global climate model. Meanwhile, the simultaneously measurements of particle number size distributions, direct CCN number concentrations as well as the chemical components of fine particles are needed in the further studies.

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Table 1. Classification statistics of events at PKU and SDZ. *N* (%): *N* is the number of each class and the value in bracket indicates the percentage accounting for the days with valid data.

Site	Class I	Class II	Non-event	Undefined	Valid	Bad/Missinga
PKU	67 (26 %)	29 (12 %)	80 (32 %)	77 (30 %)	253	22
SDZ	69 (31 %)	18 (8 %)	87 (39 %)	49 (22 %)	223	52

Table 2. Statistics of condensable vapor concentration (C), condensation sink (CS) and source rate (Q) during the NPF event over the three seasons and whole measurements period. CS is the median value during the timeframe for determination of GR.

	# Event	CS ($\times 10^{-2}$ s^{-1})		C ($\times 10^{7}$ cm^{-3})		Q ($\times 10^{6}$ cm^{-3} s^{-1})	
	PKU/SDZ	PKU	SDZ	PKU	SDZ	PKU	SDZ
Spring	29/26	2.6 ± 1.4	2.4 ± 2.1	8.7 ± 2.9	7.7 ± 2.8	2.2 ± 0.4	1.8 ± 0.6
Summer	18/10	2.2 ± 1.	0.9 ± 0.8	11.6 ± 5.7	7.9 ± 3.8	2.5 ± 0.6	0.7 ± 0.3
Autumn	20/33	1.7 ± 1.2	0.9 ± 0.6	8.1 ± 2.2	6.4 ± 3.1	1.4 ± 0.3	0.6 ± 0.2
Average in total	67/69	2.2 ± 1.3	1.4 ± 1.6	9.3 ± 4.0	7.1 ± 3.2	2.1 ± 0.5	1.2 ± 0.5

Table 3. Statistics of three quantities (CCN_{45}, CCN_{90} and CCN_{200}) at two sites and the potential contributions of anthropogenic emissions.

	PKU_average		SDZ_average		Anthropogenic emissions (%)		
	Increase number ($\times 10^{3}$ cm^{-3})	Increase factor	Increase number ($\times 10^{3}$ cm^{-3})	Increase factor	Min	Max	Average
CCN_{45}	13.0 ± 4.7	5.6 ± 3.5	7.7 ± 6.1	8.7 ± 7.6	13	91	59 ± 23
CCN_{90}	5.7 ± 2.6	6.0 ± 3.9	3.4 ± 3.1	7.0 ± 6.5	20	95	63 ± 24
CCN_{200}	1.2 ± 0.7	5.6 ± 3.7	0.8 ± 0.7	6.5 ± 6.3	10	94	62 ± 25

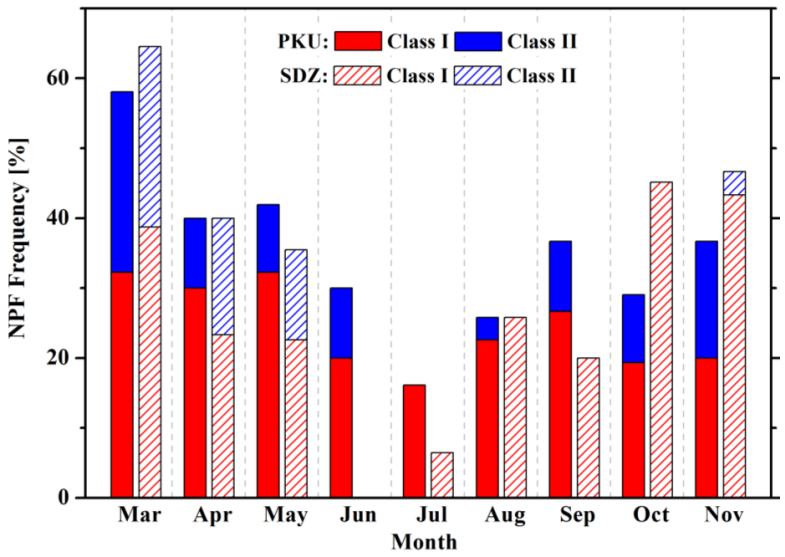

Fig. 1. The frequencies of NPF events vs. month at two sites during the whole measurement period.

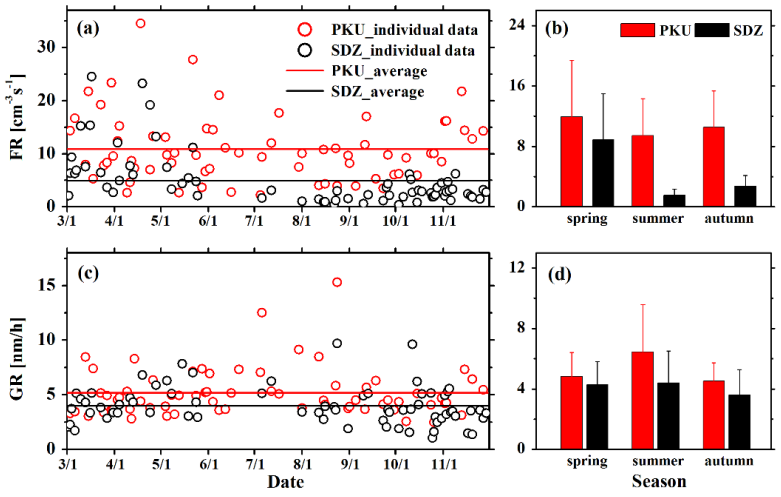

Fig. 2. The variations of particle formation rates (FR, **a** and **b**) and growth rate (GR, **c** and **d**) at PKU (red) and SDZ (black) during measurement period. The bars in **(b)** and **(d)** indicate the standard deviation.

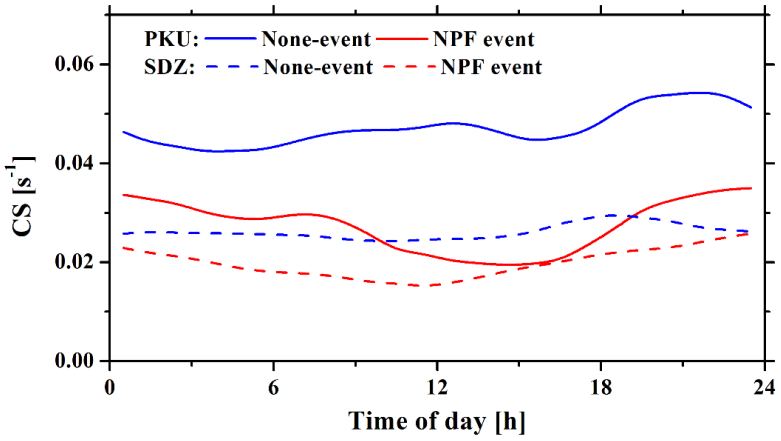

Fig. 3. Diurnal variations of condensation sink (CS) on NPF event (red) and none-event (blue) at PKU (solid line) and SDZ (dash line).

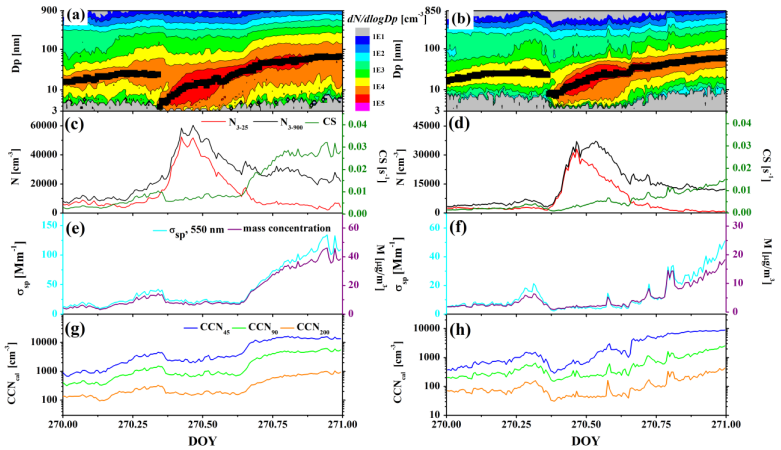

Fig. 4. (a) and **(b)**: The particle number size distributions and mean diameters of the dominating mode (black square); **(c)** and **(d)**: the evolution of condensation sink (CS, olive) and the number concentrations of nucleation mode ($N_{3–25}$, red) and total ($N_{3–900}$, black) particles; **(e)** and **(f)**: the calculated particle light scattering coefficient at 550 nm (σ_{sp}, cyan) and mass concentration (M, purple); **(g)** and **(h)**: the calculated CCN number concentration (CCN_{cal}) in certain diameter ranges (45–900 nm, blue; 90–900 nm, green; 200–900 nm, orange). Left and right panels represent the PKU and SDZ sites, respectively.