Atmospheric new particle formation in China

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Abstract. New particle formation (NPF) studies in China were summarized comprehensively in this paper. NPF frequency, formation rate and particle growth rate were closely compared among the observations carried out at different types of sites in different regions of China in different seasons, with the aim of exploring the nucleation and particle growth mechanisms. The
10   interactions between air pollution and NPF are discussed, emphasizing on the properties of NPF under heavy pollution conditions. The current understanding of NPF cannot not be fully explain the frequent occurrence of NPF at high aerosol loadings in China, and possible reasons for this phenomenon are proposed. The effects of NPF and some aspects of NPF research requiring further investigations are also summarized in this paper.

1.   Introduction

15   Atmospheric aerosols have adverse effects on human health and visibility, and cause severe air pollution in many countries (Kaiser, 2005;Cheng et al., 2011;Hand and Malm, 2007;Lelieveld et al., 2015). In addition, aerosol particles influence the Earth's radiation balance due to their direct extinction of light and by serving as cloud condensation nuclei (CCN) or ice nuclei (IN), causing very high uncertainties in predicting the ongoing climate change (IPCC, 2013). In order to understand these effects better, and especially to reduce the uncertainty of evaluating their role in climate change, comprehensive knowledge
20   about the formation and growth of aerosol particles in the atmosphere is required. Atmospheric new particle formation (NPF) is the dominant source of atmospheric aerosol particles on a global scale and has attracted broad attention for couple of decades (Kulmala et al., 2013;Kulmala et al., 2004;Merikanto et al., 2009;Dunne et al., 2016).

Generally, NPF includes the following separate steps: (1) chemical reactions in the gas phase to produce low-volatility vapor(s), (2) cluster formation from gaseous vapors nucleation, (3) nucleation or barrierless nucleation, (4) activation of
25   clusters with a second group of vapors to form a critical nucleated particle, and (5) subsequent condensational growth of nucleated particles to detectable sizes or even larger (Kulmala et al., 2014). NPF starts from atmospheric clustering. The key precursors of clusters have extremely low-volatility, including sulfuric acid (Sipila et al., 2010;Kirkby et al., 2011) and highly oxygenated molecules (HOM) (Bianchi et al., 2016;Tröstl et al., 2016;Kirkby et al., 2016;Ehn et al., 2014). Molecular clusters seem to be continuously generated almost everywhere and all the time (Kulmala et al., 2017). These clusters can be further

stabilized by reacting with other gaseous compounds like amines, ammonia and highly oxygenated molecules (HOMs), or through electrostatic interactions in the presence of ions (Kirkby et al., 2016), after which they will grow to larger nanoparticles or will be scavenged by existing surfaces. Therefore, there are two main factors controlling whether NPF will be detected in the atmosphere. One is how fast the clusters grow, while the other is how fast the clusters are scavenged (McMurry and

5   Friedlander, 1979;Kerminen et al., 2001;McMurry et al., 2005;Kuang et al., 2010). Sulfuric acid and organics are the main contributors to aerosol growth. Generally, condensation of sulfuric acid gives an important, sometime dominant, contribution to the initial growth, while organics became more and more important as the particle size is increased (Xiao et al., 2015;Kulmala et al., 2016b). High concentrations of these growth contributors will help the nanoclusters grow to sizes large enough be detected. Meanwhile, pre-existing aerosol particles act as a sink for these precursors, small clusters and newly

10   formed particles, and thereby suppress the occurrence of a NPF event.

Several gas compounds and precursors have been shown to influence NPF under conditions relevant to the atmosphere, such as SO_2/H_2SO_4 (Sipila et al., 2010), NH_3 (Kirkby et al., 2011;Kürten et al., 2016), amines (Almeida et al., 2013), VOCs/HOMs (Riccobono et al., 2014;Ehn et al., 2014;Bianchi et al., 2016), NOx (Wildt et al., 2014) and iodine species (Sipilä et al., 2016). Meanwhile, many of these compounds are responsible for secondary aerosol formation, which is very pronounced

15   during pollution episodes (Zhang et al., 2015b;Guo et al., 2014). The concentrations of these precursors and preexisting aerosol particles can both be high in polluted cities, especially in the developing countries like China and India, and cause some special features in NPF events compared with cleaner environments, which we cannot explain yet (Kulmala et al., 2017). In China, the rapid economic development and urbanization have led to high emission of various pollutants from coal combustion, motor vehicle exhaust and various industrial emissions, and resulted in highly complex air pollution. Besides high concentrations of

20   fine particles ($PM_{2.5}$, particulate matter with diameter less than 2.5 µm), high concentrations of SO_2, NO_x, NH_3, and volatile organic compounds (VOCs) were observed in frequent haze pollution episodes (Liu et al., 2013;Ye et al., 2011;Zou et al., 2015;Wang et al., 2015a). Due to a large proportion of energy supply from coal combustion, the concentration of SO_2 was thought to be the highest in the world (Bauduin et al., 2016), with surface concentrations in the range of a few ppb to over 100 ppb in north China (Sun et al., 2009;Li et al., 2007). The emissions and concentrations of SO_2 were decreasing in most regions

25   of China during the recentZu years (Lu et al., 2010;Wang et al., 2015b), but high concentrations (dozens of or over 100 ppb) of SO_2 are still being frequently observed during the heating period in winter (Wang et al., 2015a;Zhang et al., 2015a). Unlike SO_2, emissions of NOx are also highly related to traffic. NOx emissions in China showed a decreasing trend from 2012 onwards, which was later than that observed for SO_2 (Ronald et al., 2017). Several studies have found that high $PM_{2.5}$ concentrations are strongly associated with the increasing concentrations of NOx (Wang et al., 2013a;He et al., 2014;Ma et al., 2018;Sun et

30   al., 2016). NOx concentrations usually range from a few ppb to dozens of ppb in Chinese cities, while during severe haze pollution episodes NOx concentration in the city center can be even higher than 300 ppb (He et al., 2014;Sun et al., 2016). For the most important alkaline gas, i.e. NH_3, there has been no national scale measurement in China despite its extensive emissions

and increasing emission trend (Fu et al., 2015). High concentrations of NH_3 (maximum concentration higher than 100 ppb) (Meng et al., 2015b;Wen et al., 2015) and strong correlations between the peak levels of fine particles and large increases in NH_3 concentrations (Liu et al., 2015;Ye et al., 2011) were observed in the North China Plain. Unlike SO_2, emissions of NH_3 are mainly from non-point sources difficult to control. Emission of VOCs have a similar situation as NH_3. The total emissions

5 of VOCs in China were estimated to be still increasing during the recent years (Wei et al., 2011;Wu et al., 2016). Observation data showed that the annual average mass concentration of total non-methane hydrocarbons (NMHCs) was about 10^2 μg m^{-3}, or dozens of ppb in urban and suburban site in Chinese cities, which is much higher than that in North America (Zhang et al., 2017a;von Schneidemesser et al., 2010;Parrish et al., 2009;Zou et al., 2015). High concentrations of these gas precursors have resulted in high concentrations of secondary inorganic and organic species in $PM_{2.5}$ during haze formation (Yang et al.,

10 2011;Zhao et al., 2013;Dan et al., 2004;Duan et al., 2005;Wang et al., 2012), but how this cocktail of high concentrations of SO_2, NOx, NH_3, VOCs and particulate matter (or highly complex air pollution) influence NPF remains highly uncertain.

Atmospheric NPF has been observed globally, including in almost all kinds of environments (Kulmala et al., 2004;Wang et al., 2017;Kulmala et al., 2016b;Manninen et al., 2010;Nieminen et al., 2018;Kerminen et al., submitted). However, no uniform theory has been found that would explain the occurrence and characteristics of NPF in different atmospheric

15 environments. Generally, NPF was observed less frequently than expected in pristine environments, while more often than theoretical prediction in polluted cities (Kulmala et al., 2017). In this study, we will summarize the NPF studies conducted in polluted regions in China, focusing on the properties of these NPF events, and trying to figure out the possible reasons for the frequent occurrence of NPF at high aerosol loadings. Recently, Wang et al. (2017) summarized the techniques, recent advances, current bottlenecks and future directions in studying NPF in China, while this study will provide a more comprehensive

20 summary of the characteristics of NPF and will emphasize the interactions between air quality and NPF.

2. Overview of NPF research in China

Field observation related to atmospheric NPF started around 2004 in China (Wu et al., 2007). After that, observations concerning NPF were carried out in North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD), western China cities like Lanzhou, Xi'an and Urumqi, and coastal cities as well as adjacent seas. NCP, YRD and PRD are the most

25 developed regions in China and they all have a high intensify of population. The air pollution level decreases from NCP to PRD, or from north to south, among these three regions. The western China cities like Xi'an and Urumqi suffered by heavy air pollution. Xi'an was reported to have a much higher fine particle concentration than Beijing in NCP (Huang et al., 2014).

A map of observation stations involving NPF study in China is shown in Figure 1. These observation sites include urban and suburban sites like Beijing, Shanghai, Nanjing, Guangzhou etc., regional and rural sites like Shangdianzi, Yufa, SORPES

30 (Nanjing University), Backgarden, Kaiping, etc., and mountain sites like Waliguan, Tai, Heng, Huang etc., providing

information on aerosol size distribution in different environments. Besides routine observations, comprehensive campaigns like PRIDE-PRD2004, CAREBeijing-2006, and CAREBeijing-2008 were also carried out for better understanding of NPF and aerosol pollution in representative region and period in China. Long-term observations on NPF are relatively rare in China, and only a few studies reported NPF observations covering longer than a one-year period (Wu et al., 2007;Kivekas et al.,

5   2009;Yao et al., 2010;Wu et al., 2011;Shen et al., 2011;Qi et al., 2015;Peng et al., 2017).

Up to now, about 100 papers from about 20 groups have been published related to NPF in China. Most of these studies focused on the characterization of NPF events, such as the properties and time evolution of the particle size distribution, particle formation and growth rate, condensation sink, etc. Some of them also studied the favorable conditions for NPF, including the influences of relative humidity (RH), temperature, wind speed and direction, air mass origin etc. Few of these studies

10   investigated NPF mechanisms involving the nucleation participants, the growth contributors and the scavenging process by preexisting aerosols, while some others investigated various effects of NPF, especially the contribution of NPF to atmospheric CCN.

The aerosol size distribution and its time evolution provide the basic information for studying NPF. Many studies about NPF in China measured only particles larger than 10 nm, while a few studies also measured particles with diameters in the

15   range of 3-10 nm. However, the information of particles smaller than 3 nm is quite limited due to the lack of detection instruments, and only few studies reported measurement results of sub-3 nanoparticles (Xiao et al., 2015;Cai et al., 2017;Cai and Jiang, 2017;Jayaratne et al., 2017). As for gas phase precursors, direct measurements of H_2SO_4 were carried out with atmospheric pressure-ion drift-chemical ionization mass spectrometer (AP-ID-CIMS) in a few studies (Yue et al., 2010;Zheng et al., 2011;Wang et al., 2011), while other studies usually estimated H_2SO_4 concentrations using different proxies related to

20   SO_2, radiation, O_3, and relative humidity (RH). Amines and ammonia are crucial in NPF since they can stabilize sulfuric acid clusters by forming acid-base complexes, but very little NPF research measured these compounds in China (Zheng et al., 2015b). Research on other relevant gaseous compounds like HOMs, or on air ions, is also very limited in China yet. Comprehensive, long-term and high quality relevant measurements are required for a better understanding about the nucleation and growth mechanisms of nanoparticles in China.

25   **3.   Characterization of NPF events in China**

There are a few basic parameters to characterize NPF events, which are listed in Table 1. Most of the NPF researches in China calculated these parameters, as listed in Table 2. In the following chapters, we will summarize and discuss the measurement results of the frequency of NPF events, new particle formation rate (FR), particle growth rate (GR) and the related concentrations and source rate of condensation vapors. Although there were differences in calculating these parameters by

30   different groups, we will not discuss much about the methodology since the main purpose of this paper is to provide an

overview of the characteristics of NPF in China.

3.1 NPF frequency

The primary information in studying atmospheric NPF is whether it is taking place or not, i.e. to identify NPF events. Unfortunately, there is no unique mathematical criterion or definition for a NPF event. Dal Maso et al. (2005) suggested criteria

5 for justifying a NPF event: a distinctly new mode of particles start in the nucleation mode size range, prevails over a time span of hours, and shows signs of growth. The particle growth is important for separating a NPF event from particles associated with local emission sources like traffic, especially when the particle size detection limit of the instruments is not low. In addition to NPF event days, the days with an absence of particles in the nucleation mode size range are called non-event days. However, some days are not easy to be classified as either events or non-events, so they usually classified as undefined days.

10 Most NPF studies in China used similar methods, but certainly subjective biases existed. In this paper, as mentioned earlier, we will not pursue the details of the justifying methods, but focus on the statistic results of the measurement.

NPF events were observed with quite different frequencies ranging from less than 10% to more than 50% in different environments and different seasons. In Figure 2, we summarize the reported NPF event frequencies in China according to the season, observation site type and region, but ignoring observations of too short period like less than one month. Generally, low

15 frequencies were observed in remote clean environments like at mountain sites (Zhang et al., 2017c) and on marginal seas (Liu et al., 2014), while there were no significant differences among urban, suburban and rural or regional sites. Although higher NPF event frequencies were sometimes observed in an urban site compared with a rural site in the same region (Yue et al., 2013), NPF was usually found to be a regional phenomenon in China. For example, NPF in the Beijing urban area always coincided with NPF at a regional site 120 km away (Wang et al., 2013c). Despite the similar frequencies, much higher FR (by

20 220%) and GR (by 50%) were observed in the Beijing urban site than in the corresponding regional background site (Yue et al., 2009;Wang et al., 2013c). The corresponding values of Q, Ccv, and CS were also larger at Beijing than those at the regional site Yufa by 40%, 40%, and 60%, respectively (Yue et al., 2009). These results indicated that the higher pollution level in Chinese cities usually resulted in stronger NPF events compare to rural areas. As for different regions, there seemed to be no significant differences in the NPF event frequency between NCP, YRD and western China cities. PRD had a relatively lower

25 NPF frequency compared with these three regions, but the difference was not statistically significant.

NPF event frequencies were also different between the different seasons. In north China, spring is usually the season with the highest frequency of NPF events, which is probably due to the typically low condensation sink (CS), relatively high solar radiation intensity, and low temperature and RH (Shen et al., 2011;Wu et al., 2007). In the NCP of China, many studies observed that summer had the lowest NPF event frequency although the condensable vapor concentration was the highest

30 during summer months due to enhanced photochemical process (Shen et al., 2011;Wu et al., 2007;Yue et al., 2009). The lowest

frequency of NPF events during summertime in NCP might be related to the high temperatures and RH, together with the stagnant and polluted air masses which could cause a high CS (Wu et al., 2007). In the YRD region, high NPF event frequencies were observed in spring and summer, although the temperature and RH were high in summer (Zhu et al., 2013;Qi et al., 2015). A low temperature favors NPF (Zhu et al., 2013), but according to the our summary, as shown in Figure 2, low NPF event

5   frequencies were usually observed in winter, which might be due to the weak solar radiation as well as typically high pollution levels at that time of the year.

Air pollutants and meteorological features are usually studied together with nanoparticles and their precursors. By comparing the pollution character between NPF events and other days, the primary factors affecting NPF events might be identified. Cai et al. (2017) found that the Fuchs surface area (which is a representative parameter of coagulation scavenging based on kinetic theory and is proportional to CS) fundamentally determined the occurrence of NPF events. The Fuchs surface

10   area had a good correlation with the $PM_{2.5}$ mass concentration, and no NPF event was observed when the daily mean $PM_{2.5}$ concentration was higher than 43 μg m^{-3} in the winter of 2015 in Beijing (Jayaratne et al., 2017). However, in some cases, the CS or the average coagulation sinks during NPF events were not significantly lower compare to other times when new particles were not formed, indicating that other factors, such as the precursor vapors and photochemical activity, might also play an

15   important role in driving NPF (Gong et al., 2010).

The NPF event frequency can be quite different in air masses from different directions (Wu et al., 2007). Higher NPF event frequencies were usually observed within relatively clean air masses having a low CS (Zhu et al., 2013;An et al., 2015;Jayaratne et al., 2017;Peng et al., 2017). However, in some cases, NPF events also occurred in polluted air masses. For example, during the summer in Beijing, NPF was observed under low wind speed conditions and this phenomenon usually

20   coincided with a wind direction change from north to south, where the air is more polluted (Zhang et al., 2011). Similarly, in Hong Kong NPF was usually observed when air masses originated from the northwest to northeast directions (Guo et al., 2012). At the summit of Mt. Tai, continental air mass passing through more polluted areas also favored NPF (Lv et al., 2018). Consecutive NPF events were observed in the presence of strong biomass burning plume at a downwind rural site in PRD (Wang et al., 2013e). Meanwhile, compare to the NPF events taking place in clean air masses, the FR seemed to be lower and

25   the GR seemed to be higher in the NPF events taking place in a polluted air mass plume (Qi et al., 2015). An observation in North China Plain reported that when the air mass was transported from the polluted south area, the average PM_{10} (PM with diameter less than 10 μm) concentrations in NPF event days were higher than during the non-event days (Shen et al., 2011). In addition, air masses from polluted north China favored the occurrence of regional NPF, while clean air mass from east usually caused local NPF in Nanjing in YRD region (Dai et al., 2017).

30   Besides the condensation sink, meteorological factors and air mass directions, NPF events seemed not to be very sensitive to the concentration levels of common gas pollutants in China, such as O_3, SO_2, and NO_2 (Zhu et al., 2013;An et al., 2015). It was observed that SO_2 concentrations were lower during the NPF event days than during non-event days in NCP (Herrmann

et al., 2014) and Taiwan (Young et al., 2013a) as well as during autumn and winter in YRD (Qi et al., 2015), whereas higher

SO_2 concentrations on NPF days were only observed during spring and summer in YRD (Qi et al., 2015;Yu et al., 2016), during

autumn in PRD (Gong et al., 2010), and at mountain sites (Zhang et al., 2017c). Meanwhile, based on the empirical parameter

developed to judge whether NPF will occur or not, the exponent of SO_2 in this empirical parameter was quite small, indicating

5   there is usually enough SO_2 for NPF to occur under heavily polluted conditions (Herrmann et al., 2014). Similar results for

sulfuric acid were reported and it was found that sulfuric acid concentrations were not significantly higher (even lower,

sometimes) on NPF days compared with non-event days (Qi et al., 2015;Xiao et al., 2015;Cai et al., 2017). Overall, the previous

results seem to suggest that SO_2 was not a limiting factor for NPF in China, and similar conclusion might also be made for

sulfuric acid. However, NPF might have different patterns in environment with abundant SO_2 or not. More strong nucleation

10   but weak growth of particles were observed with high concentrations of SO_2 in polluted air masses characteristic of urban

(heavy traffic emission) or power-plant plumes, in spite of similar CS with lower concentrations of SO_2 (Gao et al., 2009;Yue

et al., 2010).

3.2 Formation rate

Due to the lack of measurements down to particle diameters of about 1.5 nm, most atmospheric nucleation rates were inferred

15   indirectly only by measuring the particle formation rate at some larger size in most of the NPF studies in China. The FR at

larger sizes (the "apparent" particle formation rate) can be related to the FR of critical clusters (the "real" nucleation rate) by

Kerminen–Kulmala equation and its revised version (Lehtinen et al., 2007;Kerminen and Kulmala, 2002), but the nuclei GR

and coagulational scavenging rate (CoagS or CS) are needed. In this study, we did not convert the "apparent" particle formation

rate into "real" nucleation rate due to the lack of GR or CS information in many published studies, but summarized FR

20   calculated at different particle sizes (Figure 3). The observed FR ranged from less than 0.1 cm^{-3} s^{-1} at particle sizes larger than

10 nm to more than 10^3 cm^{-3} s^{-1} at particle sizes below 2 nm. At a certain particle size, the FR could still differ by 2 orders of

magnitude due to the different environmental conditions. For example, many studies reported the FR of 3 nm particles ranging

from less than 1 to several tens of cm^{-3} s^{-1}.

Due to the wide range of FR under different environmental conditions, it is not easy to determine differences in FR

25   between different site types, regions or seasons. In principle, a higher CS causes a more rapid scavenging of clusters and small

particles, resulting in lower FR (Zhu et al., 2014;Man et al., 2015). According to the equation developed by Herrmann et al.

(2014) based on observation date in YRD region, FR is also inversely proportional to the CS. However, when NPF was studied

in an urban site and regional site at the same time, FR was usually higher at the urban site in spite of the higher CS, indicating

much more abundant precursors for NPF in the polluted urban environment (Wang et al., 2013c). As for NPF at a same

30   observation site but in different seasons, the highest FR was observed in summer in NCP (Shen et al., 2011) and in spring in

YRD (Qi et al., 2015).

Although the nucleation mechanism in different environmental conditions remains unknown according to current knowledge, neutral clusters of sulfuric acid, stabilized with additional vapours such as ammonia, amine, HOM should play a key role in NPF (Kulmala et al., 2014;Kulmala et al., 2013). A positive relationship between FR and the sulfuric acid

5 concentration (or H_2SO_4 proxy) was observed in many NPF studies in China. The fitted exponent between FR and sulfuric acid concentration ranged from 0.65 to 2.4 (Cai et al., 2017;Xiao et al., 2015;Dai et al., 2017), while sometimes even higher values between 2.5 and 7 were found (Wang et al., 2011). These exponents were observed to increase with an increasing CS in Beijing (Wang et al., 2011). Besides sulfuric acid, also organics, NH_3 and amines were found to be important in atmospheric particle nucleation (Wang et al., 2015c). As we mentioned earlier, although CS was much higher at urban sites, the FR was

10 usually higher at corresponding regional sites (Wang et al., 2013c). Meanwhile, SO_2 is a regional pollutant and its concentrations were similar between regional sites and city area (He et al., 2014;Ma et al., 2018). These features indicate important roles of other gas precursors in NPF in the air pollution complex of China. In fact, some observations showed that the correlation between FR and NH_3 was better than that between FR and H_2SO_4 (Xiao et al., 2015). Dai et al.(2017) proposed that plumes containing high concentrations of ammonia, amines or HOMs produced from their observed VOCs led to strong

15 local NPF events. Recently, Yao et al. (2018) reported a long-term continuous observations for NPF in urban Shanghai and observed one to two orders of magnitude higher FR than typical values in the clean atmosphere. These observed FR were far higher than those derived from H_2SO_4–H_2O or H_2SO_4–NH_3–H_2O mechanisms but close to those observed in the H_2SO_4–DMA–H_2O laboratory experiments, and coincided with sulfuric acid clusters and sulfuric acid–dimethylamine (DMA) clusters. These results suggested that H_2SO_4–DMA–H_2O nucleation played important roles in the NPF Chinese megacities. Up to now, there

20 is still quite limited investigations for the relation between FR and organics, NH_3 and amines in China and it is certainly crucial for a better understanding of NPF in polluted area. Ion-induced of pure organic nucleation was proposed to be important according to chamber experiments (Kirkby et al., 2016), but seems to have a minor role in the polluted environment in China (Herrmann et al., 2014;Xiao et al., 2015;Yao et al., 2018). This is understandable because the ion production rate is usually much lower than FR in China.

25 **3.3 Growth rate**

Growth of nano-particles is crucial for NPF. The GR determines the size that new particles can grow to before being scavenged, i.e. a higher GR results in a larger particle diameter (Zhu et al., 2014;Man et al., 2015). There are several methods to calculate GR from the time variations of particle size distributions, such as the appearance time method (Kulmala et al., 2013) and mode fitting method (Kulmala et al., 2012), or by solving the general aerosol dynamics equation (Pichelstorfer et al., 2018).

30 Regardless of the possible difference caused by using different calculation methods, GRs reported in China varied a lot

from an urban area to a rural region and from spring to winter, ranging from a few nm h[-1] to more than 20 nm h[-1] (Figure 4). Generally, GRs in urban sites were found to be higher than in their regional sites (Wang et al., 2013c). This is probably caused by the more abundant condensing vapors in polluted cities, although there are limited data on sulfuric acid and low-volatile organic vapor concentrations in China. No significant difference were found among the observations carried out in different

5 regions in China in spite of the different pollution levels. Lower GR was observed at a mountain site compare to that in an urban area (Wang et al., 2014), but the GR of large-size particles in mountain site could be as high as about 10 nm h[-1] (Nie et al., 2014;Wang et al., 2014). For GRs in different seasons, higher GRs were observed in summer than other seasons, indicating higher concentrations of condensable vapors, which may be related to strong photochemical and biological activities (Zhu et al., 2013;Shen et al., 2011;Qi et al., 2015). GR is also dependent on the concerned size of the particles, larger particles usually

10 having a larger GR, which could also be inferred from the data summarized in Figure 4.

Sulfuric acid and organic vapors with low volatility were thought to be the main contributors to the growth of particles formed by NPF. Generally, sulfuric acid was thought to be the dominant contributor for the growth of newly formed particles, but became less and less prominent for the growth of larger particles (Xiao et al., 2015). For example, Xiao et al. (2015) and Yao et al. (Yao et al., 2018) calculated and estimated that sulfuric acid was enough to explain the observed growth for particles

15 smaller than 3 nm, but was insufficient to explain the observed growth rates of large particles. They further calculated the relative contribution ratio of sulfuric acid to the particle growth in different particle size ranges, being 39% for 2.39-7 nm and 29% for 7-20 nm in urban Shanghai (Xiao et al., 2015), 3% to 14% for 7-30 nm in urban Beijing (Wang et al., 2015c), about 26 % for 5.5-25 nm in suburban Hong Kong (Guo et al., 2012), and about 29% during the Beijing Summer Olympic period (Gao et al., 2012). Some studies reported that H_2SO_4 had a negligible contribution to the growth of particles larger than 10 nm

20 (Meng et al., 2015a;Liu et al., 2014). The particle shrinkage (reversal in growth of particles size) was reported in a few studies in China. The particle shrinkage could be due to measuring particles present in different air masses during different times of the day, or the evaporation of water and/or semi-volatile species in the particles. If the air masses did not vary significantly, a similar shrinkage rate to the growth rate in the NPF events might indicate a notable fraction of semi-volatile species contributed to the growth (Young et al., 2013b;Yao et al., 2010), which is consistent with that organics being the main contributor to the

25 large-size particle growth. Yu et al. (2016) estimated that a high concentration of extremely low volatility organic compounds was the key factor leading to high GR for very small particles (1.4-3 nm) in urban Nanjing. However, Yue et al. (2010) proposed a dominant role of sulfuric acid in the growth of new particles in sulfur-rich NPF events. A model simulation study about NPF in Beijing also supported that only small fraction of organics contributed to the growth of new particles, and these organics were mainly O_3 initiated (Wang et al., 2013b). Besides sulfuric acid and organics, some studies reported a two-stage growth

30 of new particles in China, in which sulfuric acid and organics contributed to the first-stage growth in daytime while NH_4NO_3 and organics possibly contributed to the second-stage growth at nighttime (Zhu et al., 2014;Man et al., 2015;Liu et al., 2014). Tao et al. (2016) observed high level of aminiums, and suggested that the heterogeneous uptake of amines in acid-base reactions

could effectively contribute to particle growth in NPF events.

4. NPF under heavy air pollution

The heavy air pollution makes China quite a different environment for NPF compared with western countries (Wang et al., 2017;Kulmala et al., 2017;Yu et al., 2017). Generally, concentrations of particles and condensable vapors in Chinese cities and

5   regional background area are much higher in China than that in North America or Europe (Shen et al., 2016b;Wang et al., 2013c;Gao et al., 2009). The CS and small molecular cluster and particle (1–3 nm) concentrations are about an order of magnitude higher in China compared with European cities (Kulmala, 2015;Kontkanen et al., 2017). The occurrence frequencies of NPF events in high aerosol-loading environments of China were higher than those in low aerosol-loading environments (Peng et al., 2014). Meanwhile, the observed FR was much higher, and the GR was also higher (but to a less

10   great extent relative to FR) for NPF in China than that at rural/urban sites in western countries (Shen et al., 2016b). As pointed out by Cai et al. (2017), previous FR calculation may still underestimate the real nucleation rate due to underestimation or omission of coagulation among particles in the nucleation mode with strong nucleation in China.

The influence of heavy air pollution on NPF might be identified by studying NPF in periods with short-term strong air pollution control. Shen et al. (2016c) investigated NPF during the Olympics in 2008 and during APEC meeting in 2015 in Beijing. They found that a higher NPF event frequency coincided with the improved air quality during these important events

15   associated with temporary intense air pollution control actions compared to similar time of the year during 2010-2013. In spite of more frequent NPF events (Yue et al., 2010;Zhang et al., 2011), the strength of NPF decreased during these periods with temporary intense air pollution control actions, characterized with lower FR and GR values (Shen et al., 2016c). Due to the decreasing strength of NPF and also the reduction of primary emission source of fine particles, the number concentration of

20   particles decreased in spite of increased frequency of NPF. The mean number and volume concentrations of particles decreased by 41% and 35%, respectively, in August 2008 during the Beijing Olympic compared with 2004-2007 (Wang et al., 2013d). However, these temporary intense air pollution control actions had a much smaller influence on Aitken mode particles than on accumulation mode particles, according to the observations carried out during the APEC meeting in 2015 in Beijing (Du et al., 2017).

25   NPF was observed more often in high aerosol-loading environment of China than we would expect based on the current understanding of nucleation and particle growth (Peng et al., 2014;Kulmala et al., 2017). With much higher CS values in China than that at European and American sites, the difference of GR was not very obvious at the same site types between China and other countries (Peng et al., 2014). It turned out that NPF occurred frequently in megacities in China when the ratio of CS (10^{-4} s^{-1}) to GR (nm h^{-1}) was above 200, whereas it only occurred when this same ratio was less than 50 under clean and moderately-

30   polluted conditions (Kulmala et al., 2017). As shown in Figure 5, most of the observation data reported ratios of CS (10^{-4} s^{-1})

to GR (nm h^{-1}) between 200 and 500, while a few less than 200 but always higher than 50. More importantly, many studies reported NPF to take place with this ratio higher than 500 at urban and suburban sites. Such NPF events were able to take place in all regions in China (NCP, YRD, PRD and Western China), and during both winter and summer seasons.

NPF mainly occurred when the PM$_{2.5}$ concentration (CS) and gas pollutant concentrations, such as NO$_2$, CO and SO$_2$,
5   were both low (Wu et al., 2007;Dai et al., 2017;Yu et al., 2016). These gas pollutants were mainly from primary combustion emission (De Gouw and Jimenez, 2009), whereas PM$_{2.5}$, the main cause of haze, originated from both primary emission and secondary formation and the latter was thought to dominate during haze events in China (Yang et al., 2011;Zhao et al., 2013;Dan et al., 2004;Duan et al., 2005;Wang et al., 2012). NPF was found to concentrate on days with low RH in previous NPF studies in Beijing (Wu et al., 2007;Yue et al., 2009). The possible reason for this would be that photochemical reactions
10 are faster on sunny days with strong solar radiation and low RH. On the contrary, haze usually occurs at high RH when multi-phase processes would contribute more to the aerosol mass (Sun et al., 2010;He et al., 2014;Zheng et al., 2015a;Cheng et al., 2016;Liu et al., 2017) and the hygroscopic aerosols would contribute more to light extinction compared with low-RH conditions (Shi et al., 2014;Shen et al., 2015). NPF and haze are either purely secondary processes or dominated by secondary pollution processes, so there might be some common properties or internal relations between them. With this in mind, besides
15 the possible inaccurate estimation for the GR and CS as pointed out and estimated by Kulmala et al. (2017) and further discussed in detail by Yu et al. (2017), several other possible reasons might also be related to the frequent occurrence of NPF under heavy air pollution in China.

Secondary aerosols, including sulfate and organic aerosols, are still underestimated in current air quality models (Xiao et al., 2015;Chen et al., 2016;Hodzic et al., 2010), indicating unknown chemical and physical processes important for secondary
20 aerosol formation (Kulmala et al., 2014). These processes might create oxidants or change the surface properties of aerosols, and thereby limit their ability to take up condensable vapors and cause more frequent NPF (Kulmala, 2015). The effects of a high percentage of inorganic aerosol particles on the effectiveness of CS in unknown and may need to be investigated in laboratory experiments. NPF was observed during dust episodes in China, and both FR and GR were enhanced under dust conditions, indicating photo-induced, dust surface-mediated reactions might be important by producing condensable vapors
25 for NPF (Nie et al., 2014;Xie et al., 2015;Kulmala et al., 2017). Heterogeneous photochemical processes inducing new particle formation and growth might happen in the real atmosphere and need to be further investigated. In addition to these, high concentrations of sulfuric acid (10^6 molecules cm^{-3}) were observed at nighttime, indicative of non-photochemical OH sources (Zheng et al., 2011). The contribution of oxidation of SO$_2$ by Criegee radicals (Welz et al., 2012;Mauldin et al., 2012), which are generated in reactions of alkenes and ozone, and other possible surface-mediated reactions that lead to the formation of
30 nighttime sulfuric acid under complex air pollution conditions in China, need to be figured out as well.

5. Significance and future research directions for NPF study

The effects of NPF on air pollution and human health are crucial but highly uncertain. The NPF leads directly to a burst of small nanoparticles and increases the particle number concentration prominently. While NPF usually tends to occur in clean days with low CS, particle number concentrations are usually much higher on NPF event days than on non-event days (Shen

5 et al., 2016a;An et al., 2015). Kulmala et al. (2016a) studied separately nucleation, Aitken and accumulation mode particle number concentrations in Nanjing in YRD regions of China, and estimated that the majority of the particles were of secondary origin in all the modes. NPF was found to be an important influential factor on atmospheric aerosol number size distribution from remote mountains to polluted cities (Du et al., 2012;Shen et al., 2016a;Zhang et al., 2017c). NPF also changes the surface and volume size distribution. An et al. (2015) observed that NPF events had a large effect on Aitken- and nuclei-mode particle

10 surface and volume concentrations, while having limited contributions to accumulation- and coarse-mode particles. NPF was observed to increase the proportions of NH_4^+, SO_4^{2-}, NO_3^-, K^+ and Mg^{2+} in nucleation- and Aitken-mode particles compared with those in the total aerosol. Zheng et al. (2011) found that the calculated condensation rate of H_2SO_4 correlated with the Aitken-mode sulfate mass concentration but not with the accumulation-mode sulfate mass concentration.

With high concentrations of condensable vapors, newly-formed particles have a potential to grow quickly, which results

15 in an increase of PM volume or mass concentrations. In an episode with consecutive NPF events in the presence of strong biomass burning in PRD, the aerosol volume concentration increased 6.1 by mm^3 cm^{-3} in volume mass concentration per day or about 10 μg m^{-3} per day in mass concentration, with organics and sulfate accounting for 42% and 35%, respectively, of the particle mass concentration (Wang et al., 2013e). Furthermore, it was estimated that primary emissions and secondary formation provided 28 and 72% of particle number concentration and 21 and 79% of mass concentration, respectively.

20 Similarly, Shen et al. (2011) observed that about 20% of the NPF events led to a measurable increase in the particle mass concentration, with an average growth rate of about 4.9 μg m^{-3} h^{-1} for PM_1 (PM with diameter less than 1 μm) during the period of the mass concentration increase. Guo et al. (2014) reported a case with NPF followed by a continuous growth and appearance of haze pollution in Beijing, and proposed that the NPF probably led to haze formation.

In summary, we think that the effect or contribution of NPF to haze formation is still an open question. This question

25 might be difficult to be identified using only field observations. It might be crucial to develop some laboratory experiments and model simulations to help answering this question. In addition, interactions between NPF, pollution and meteorological conditions should be studied further. Heavy pollution could have significant feedbacks to meteorological conditions in China. For a case study in YRD, it was calculated that air pollution resulted in a decrease in the solar radiation intensity by more than 70 %, decrease in the sensible heat by more than 85 % and temperature drop by almost 10 K (Ding et al., 2013). These effects

30 resulted in a decrease of the boundary layer height, which further increased PM concentrations, forming a feedback loop (Petaja et al., 2016). On the other hand, NPF occurring in a free troposphere may have a major impact on the marine boundary layer

particle concentrations due to the subsidence (Clarke et al., 1998;Lin et al., 2007). When the aerosol loading was high, the distance of NPF peak above the planetary boundary layer became larger (Quan et al., 2017). These interactions would be also crucial for predicting NPF and air quality and for identifying the contribution of NPF to air pollution. Rather than ground observations, multi-dimensional measurements may need to be carried out in order to understand the atmospheric process up

5   to the free troposphere. Compared with the effect of NPF to haze formation, the health effect of high number concentrations of particles with diameters of several or tens of nanometers would be more essential. NPF usually occurs around the same time period as people commute to work. The effects of exposure to a high particle number concentration environment should be investigated.

Atmospheric nucleation and subsequent growth of newly-formed particles could have significant effects on air quality

10   and climate by contributing to CCN (IPCC, 2013). NPF were calculated to enhance CCN number significantly with ratios ranging from 1.2-1.8 in Shanghai in YRD region of China (Leng et al., 2014). Considering both NPF and non-event days, the average contributions of NPF events to potential CCN in the afternoon were calculated to be 11% and 6% at urban sites and regional sites, respectively (Peng et al., 2014). It seems that the enhancement of CCN due to NPF in China at a regional scale was larger than that in Europe (Shen et al., 2016b), which might due to the combination of a higher nucleation rate and quicker

15   subsequent condensable growth associated with higher pollution levels in China. NPF events was also found to have a greater impact on CCN in polluted urban sites than in regional or rural sites in China. For example, CCN number concentrations were observed to be enhanced by a factor of 2-6 in background regions and by a factor of 5.6-8.7 in polluted regions during the NPF event days (Wang et al., 2013c;Shen et al., 2016b). Nevertheless, the impact of NPF on the CCN number concentration was found to depend on the location and individual character of each NPF event, including different hygroscopic properties of

20   particles and thus different CCN activities during different NPF events, so Ma et al. (2016) suggested not using a fixed parameter to predict the contribution of NPF on CCN and Tao et al (2018) emphasized the importance of real-time measurements of hygroscopicity of particles.

During the past 15 years, a lot of NPF observations and related studies were carried in China but, as summarized by Wang et al. (2017), the application of state-of-the-art instruments are still quite limited in China. This limits our understanding on

25   the mechanism of NPF in different environments in China. Long periods and comprehensive observations would be the most important factor when investigating NPF mechanisms in China, while laboratory experiments and model simulations would also be very helpful and necessary. Key participants and processes of NPF under complex air pollution conditions in China still wait to be answered, and the unexpected NPF at high aerosol loadings need to be explained. Contributions of different mechanisms to NPF should be evaluated with the consideration of spatiotemporal difference, and possibly also with the

30   consideration of process of air pollution control in China. As suggested by Kulmala (2018), environmental grand challenges, such as climate change, water and food security as well as urban air pollution, are all linked and need to be studied together. The effects of NPF in China on climate change and human health is still poorly understood and should be evaluated

quantitatively. Although a global view is needed for these common challenges of mankind, densely-populated China will no doubt be a very important area in this respect. Studying these effects will be essential for future studies of NPF in China and will be important to a global effort for a better atmosphere on Earth.

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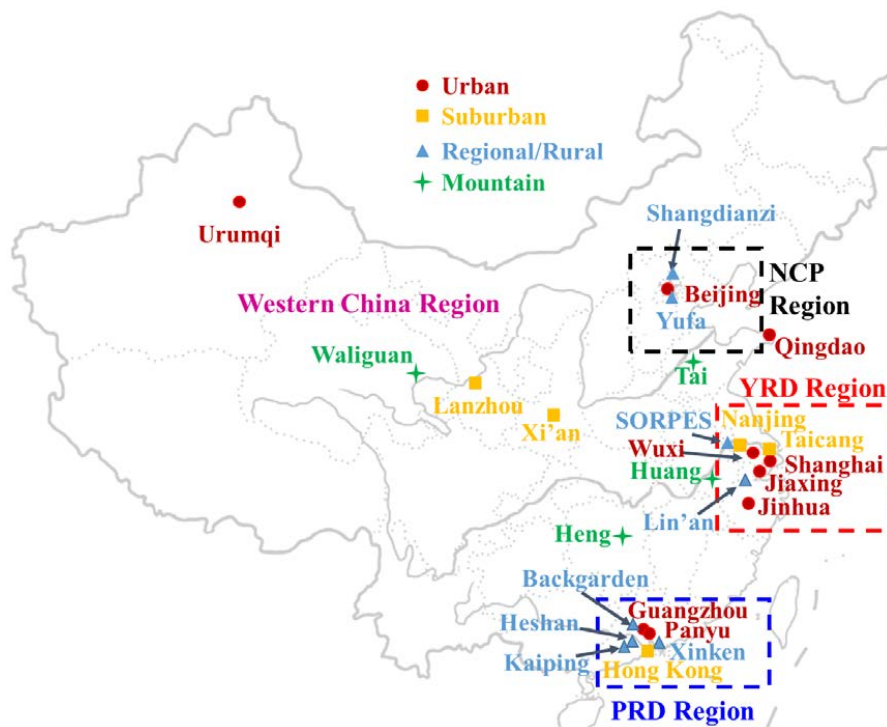
Figure 1: Map of observation sites. involving NPF study in China. Most of these observations sites were classified into four regions in this study, i.e. North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD), Western China region (Western).
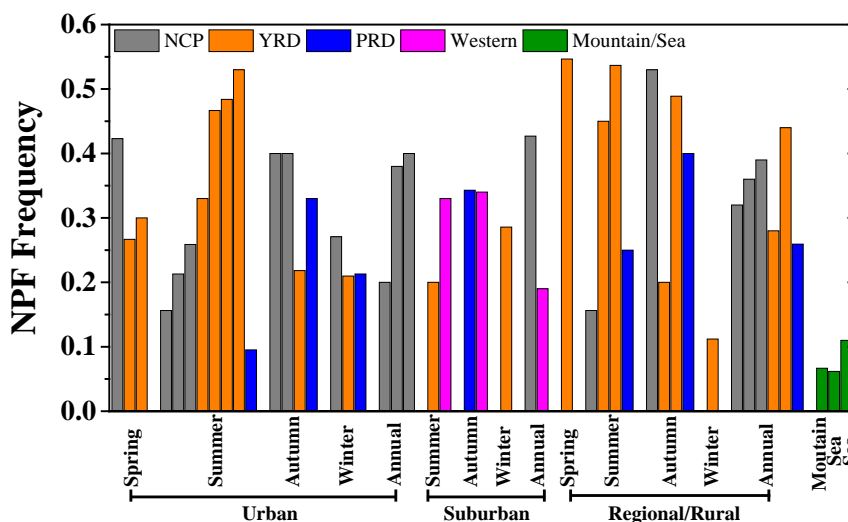
Figure 2: NPF frequency observed at different places in different seasons in China. The data are collected in the references in Table 2.
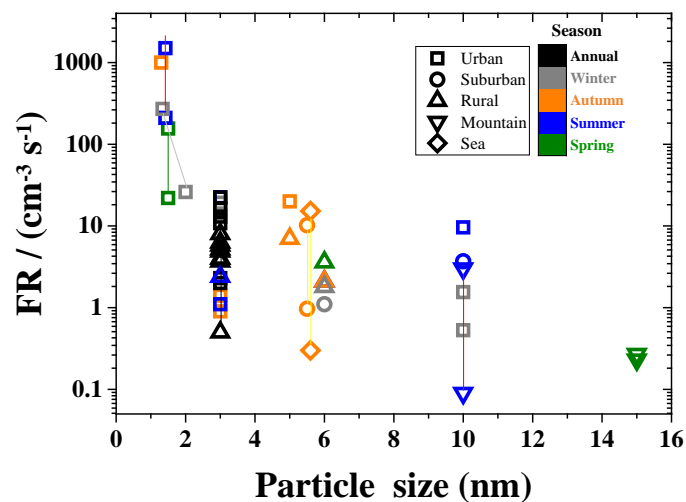
Figure 3: New particle FR observed at different places in different seasons in China. The line between two data points indicates that a range of FR was reported in the literature. The data are collected in the references in Table 2.
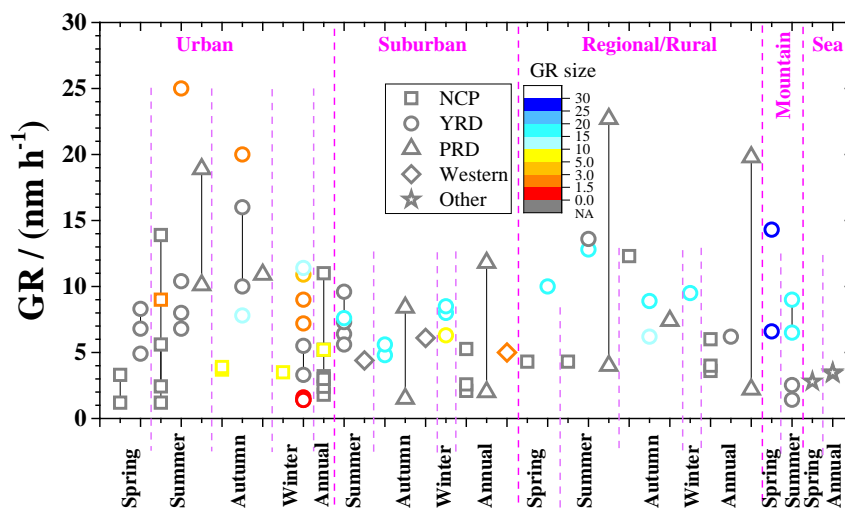
Figure 4: Particle GR observed at different places in different seasons in China. The data point are colored by the particle size range for the GR calculation. The line between two data points indicates that a range of GR was reported in the literatures. The data are collected in the references in Table 2.
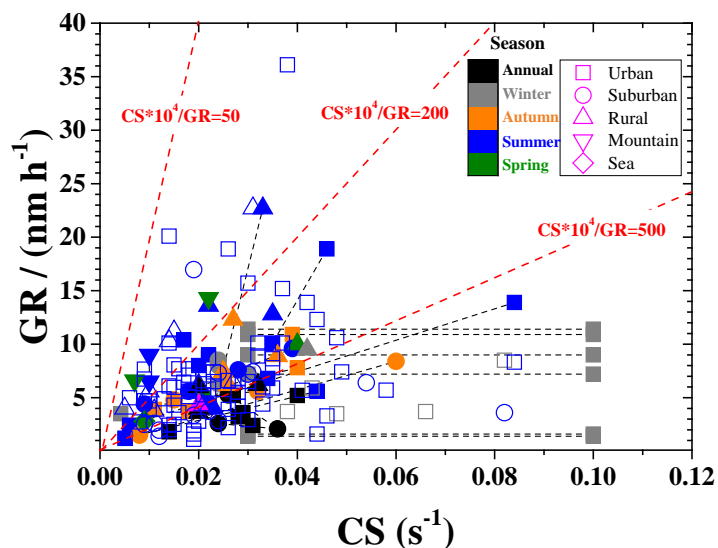
Figure 5: Particle GR as a function of CS in the NPF events in China. The solid points are average data for a certain observation period, while the open points are data for individual NPF days. The line between two data points indicates that a range of GR and/or CS was reported in the literature. The data are collected in the references in Table 2.

Table 1: Parameters to characterize NPF events

Parameter	Description	Calculated from
FR	Formation rate of particles	Temporal variation of particle size distribution
GR	Growth rate	Temporal variation of particle size distribution
CS	Condensation sink	Particle size distribution
CoagS	Coagulation sink	Particle size distribution
C_{cv}	Condensation vapor concentration	GR
Q	Source rate of condensation vapors	CS and C_{cv}

Table 2: Characterization of NPF events in China

Observation time	Observation site	Region	Site type	Season	EP*	FR (cm⁻³ s⁻¹)	Size for FR (nm)	GR (nm h⁻¹)	Size for GR (nm)	C_{cv} (10⁷ cm⁻³)	Q (10⁶ cm⁻³)	CS (s⁻¹)	Measured size (nm)	Ref.	Comments
2014.7-12 2015.6-8	Taishan	NCP	Rural	Summer Autumn	40%	7.10 (0.82-25.04)	3	1.98 (0.58-7.76)	3-20			0.014 (0.001-0.284)		(Lv et al., 2018)	
2015.8-2015.9	Beijing	NCP	Urban	Annual				3.2(1.5-6.1)** / 3.6(1.4-7.5)						(Du et al., 2017)	ground / 260m
2015.10-2016.1	Beijing	NCP	Urban	Winter	27%	26 (12-38)	2-3	3.5(0.5-9)	2-10			0.0042		(Jayaratne et al., 2017)	
2013.11-2014.12	Xi'an	Western	Suburban	Annual	19%			5±1.9	2.8-10.7				10-487	(Peng et al., 2017)	
2018.4-7	Huang	YRD	Mountain	Summer	7%	0.09-0.3	10	1.42-2.53					10-10000	(Zhang et al., 2017c)	
2014.8-11	Lanzhou	Western	Suburban	Autumn	34%	1.71		6.1					14.6-661.2	(Zhang et al., 2017b)	
2016.3-4	Beijing	NCP	Urban	Spring	42%	22-156	1.5	1.2-3.3					1-	(Cai et al., 2017)	H_2SO_4 measured
2015.9-11	Nanjing	YRD	Urban	Autumn	22%	1000 / 20	1.3 / 5	20 / 7.8	2 / 10			0.04 / 0.04		(Dai et al., 2017)	
	YRD	YRD	Regional	Autumn	20%	7	5	6.2	10			0.026			
2014 Youth Olympic	Nanjing	YRD	Urban	Summer	47%	210-2500	1.42	25	1-3				1.4-	(Yu et al., 2016)	
2013 summer	Shanghai	YRD	Urban	Summer									10-10000	(Tao et al., 2016)	
2011.11-12	Panyu, Guangzhou	PRD	Urban	Winter	21%	0.97 / 0.82 / 1.55 / 0.53	10-20	8.5 / 5.9 / 3.7 / 3.5			9.7 / 3.5 / 1.9 / 2.3	0.082 / 0.043 / 0.038 / 0.048	10-20000	(Tan et al., 2016)	

Observation time	Observation site	Region	Site type	Season	EP*	FR ($cm^{-3}\,s^{-1}$)	Size for FR (nm)	GR ($nm\,h^{-1}$)	Size for GR (nm)	C_{cv} ($10^7\,cm^{-3}$)	Q ($10^6\,cm^{-3}$)	CS (s^{-1})	Measured size (nm)	Ref.	Comments
2008.8-9	Beijing	NCP	Urban	Autumn	40%	0.57		3.7			3.4	0.066			
2010-2013				Annual	20%	0.9	3-10	3.7	3-10			0.018		(Shen et al., 2016c)	
2015.8-9				Autumn	40%	2.3		5.2				0.04			
						1.4		3.9				0.011			
2008.3–2013.12	Shangdianzi	NCP	Rural	Annual	36%	6.3(0.5-39.3)		3.6(0.7-13.4)				0.02			
2011.1–2011.12	Taishan	NCP	Rural	Annual	32%	3.7(1-9.6)	3	6(1.1-15.4)				0.02	3-800	(Shen et al., 2016b)	
2013.1–2013.12	Lin an	YRD	Rural	Annual	28%	5.8(0.8-26.5)		6.2(1.8-21.3)				0.032			
2015.5	Jiaxing	YRD	Urban	Summer	48%	9.6(4-17)	10-20	6.8(2.2-15.7)		9.4(3.0-21.5)	3.3(0.5-7.7)	0.034(0.015-0.058)	10-10000	(Shen et al., 2016a)	
2008.7-9	Beijing	NCP	Urban	Summer									3-900	(Wang et al., 2015c)	H_2SO_4, VOCs measured
2013.11-2014.1	Shanghai	YRD	Urban	Winter	21%	112.4-271	1.34	1.6±1	1.35-1.39	2.3-6.4		0.03-0.1	1.35-615	(Xiao et al., 2015)	
						2.3-19.2	3	1.4±2.2	1.39-1.46						
								7.2±7.1	1.46-1.7						
								9±11.4	1.7-2.39						
								10.9±9.8	2.39-7						
								11.4±9.7	7-20						
2011-2013	YRD	YRD	Regional	Annual	44%							0.038-0.053	6-800	(Qi et al., 2015)	
				Spring	55%	3.6	6	10	6-30			0.04-0.055			
				Summer	54%	2.1		12.8				0.035-0.05			
				Autumn	49%	2.1		8.9				0.036-0.051			
				Winter	11%	1.8		9.5				0.042-0.058			

Observation time	Observation site	Region	Site type	Season	EP*	FR (cm⁻³ s⁻¹)	Size for FR (nm)	GR (nm h⁻¹)	Size for GR (nm)	C_{cv} (10⁷ cm⁻³)	Q (10⁶ cm⁻³)	CS (s⁻¹)	Measured size (nm)	Ref.	Comments
2011	Yellow River Delta	NCP		Annual	22%	6.6		5.3						(Yuan et al., 2015)	
2012.7-8	Nanjing	YRD	Suburban	Summer		3.7(1.6-6.7)	10	7.6(5.6-9.6)	10-20	10.5(7.7-13.2)	2.9(1.9-4.7)	0.028(0.018-0.039)		(An et al., 2015)	
2010.7-8	Wuxi	YRD	Urban	Summer	53%			10.4(6.2-13.3)				0.017(0.009-0.029)		(Peng et al., 2014)	
2010.4-6	Shanghai	YRD	Urban	Summer	33%			8(4.2-12)				0.02(0.01-0.033)			
2010.11	Guangzhou	PRD	Urban	Autumn	33%			10.9(7.3-18.1)				0.039(0.026-0.056)			
2008.5-6	Urumchi	Western	Urban	Spring	86%							0.016(0.01-0.026)			
2010.6 -7	Jiaxing	YRD	Regional	Summer	45%			13.6(7.9-19.6)				0.022(0.011-0.041)			
2007.10	Yufa	NCP	Regional	Autumn	53%			12.3(8.6-21)				0.027(0.005-0.053)			
2008.10-11	Kaiping	PRD	Regional	Autumn	40%			7.4(3.2-13.5)				0.025(0.003-0.086)			
2009.10-12	BG	PRD	Sea	Autumn	15%			4.5(3.2-7.5)				0.014(0.01-0.018)			
2011.3-4	Changdao	NCP	Sea	Spring	19%			5.7(4.5-6.8)				0.02(0.019-0.021)			
2011.10-11	Wenling	YRD	Sea	Autumn	10%			7.5				0.026			
2011.3-4	China sea		Sea	Spring	11%			2.8(1.6-3.9)				0.009(0.008-0.011)			
2011.10-11, 2012.11	Marginal seas		Sea	Autumn	6%	0.3-15.2	5.6-30				1.6(1.1-2.2)			(Liu et al., 2014)	
2010.5	Qingdao	NCP	Coastal city	Summer	41%	13.3		6.2						(Zhu et al., 2014)	
2011.8-9	Huangshan	YRD	Mountain	Summer		0.25-0.67	10-20	6.5-9.0	10-20			0.01		(Wang et al., 2014)	
2011.10-11	Nanjing	YRD	Suburban	Autumn		0.83-1.67		4.8-5.6				0.015-0.032			
2011.11-2012.3	Nanjing	YRD	Suburban	Winter	29%	1.1	6	8.50 / 6.3 / 8	6-30 / 3-7 / 7-30		3.8	0.024	0.8-800	(Herrmann et al., 2014)	
2012.4	Shanghai	YRD	Urban	Spring	27%	0.40		4.91				0.021	10-800	(Leng et al., 2014)	

Observation time	Observation site	Region	Site type	Season	EP*	FR (cm⁻³ s⁻¹)	Size for FR (nm)	GR (nm h⁻¹)	Size for GR (nm)	C_cv (10⁷ cm⁻³)	Q (10⁶ cm⁻³)	CS (s⁻¹)	Measured size (nm)	Ref.	Comments
2009.4	Mountain in Hunan		Mountain	Spring		0.27	15	14.3	30-50		2.8	0.007	10-10000	(Nie et al., 2014)	dust
				Spring		0.23		6.6			1.4	0.022			non-dust
2006.7	BG	PRD	Rural	Summer	25%	2.4-4	3-25	4-22.7		5-31	1.3-11	0.023-0.033	15-10000	(Yue et al., 2013)	
	Guangzhou	PRD	Urban	Summer	10%	NA		10.1-18.9		14-26	7.7-9.1	0.035-0.046	3-10000		
2008.10-11	Kaiping	PRD	Rural	Autumn	40%			7.4 / 3.2-135.5		1.3 (H₂SO₄)		0.025 / 0.03-0.086		(Wang et al., 2013e)	
2008.3-11	Beijing	NCP	Urban	Annual	38%	10.8(2.2-34.5)		5.2		9.3	2.1	0.027		(Wang et al., 2013c)	
	Shangdianzi	NCP	Regional	Annual	39%	4.9(0.4-24.5)		4		7.1	1.2	0.02			
2010.10-11	HK	PRD	Suburban	Autumn	34%	0.97-10.2	5.5	1.5-8.4		0.8-1.2 (H₂SO₄)		0.008-0.06	5.5-350	(Guo et al., 2012)	
2010 summer		YRD	Suburban	Summer	20%									(Zhu et al., 2013)	
2009 autumn	Nanjing		Urban/Suburban	Autumn	3%			10-16							
2011 spring			Urban	Spring	30%			6.8-8.3							
2008	Beijing	NCP	Suburban	Annual	43%			3.2(1.2-8)		4.4	1.2		10-1000	(Gao et al., 2012)	
2006.6-7	Lanzhou	Western	Suburban	Summer	33%	1.8-3.4	10-20	4.4(1.3-16.9)		6.1	1.1	0.009-0.022	10-10000	(Gao et al., 2011)	
2008.10-2009.2	Shanghai	YRD	Urban	Winter	5%	0.2-0.5	10-20	3.3-5.5					10-10000	(Du et al., 2012)	
2004-2006	Beijing	NCP	Urban	Annual				0.4		0.6		0.01	3-10000	(Wu et al.,)	

Observation time	Observation site	Region	Site type	Season	EP*	FR (cm⁻³ s⁻¹)	Size for FR (nm)	GR (nm h⁻¹)	Size for GR (nm)	C_{cv} (10⁷ cm⁻³)	Q (10⁶ cm⁻³)	CS (s⁻¹)	Measured size (nm)	Ref.	Comments
										(H_2SO_4)					
2008.7-9	Beijing	NCP	Urban	Summer	26%			9	1.5-3			0.022	3-900	(Wang et al., 2011)	H_2SO_4 measured
2008.3-2009.8	Shangdianzi	NCP	Rural	Spring/Summer	36%	8(0.7-72.7)		4.3(0.3-14.5)				0.02	3-10000	(Shen et al., 2011)	
2008.6-9	Beijing	NCP	Urban	Summer	21%			2.43-13.9				0.006-0.084	12-550	(Zhang et al., 2011)	
2008	Beijing	NCP	Urban	Annual	10-70%	2-13		3-11					3-900	(Yue et al., 2010)	
2003-2004	HK	PRD	Coastal/Suburban	Annual		3.6(2.3-11.5)		2-11.8					3.2-106	(Yao et al., 2010)	
2006	Beijing	NCP	Urban	Summer	16%	1.1-22.4		1.2-5.6		1.5-17	0.1-8.8	0.0051-0.044	3-10000	(Yue et al., 2009)	
	Yufa	NCP	Rural	Summer	16%										
2005.5-6	Taicang, YRD	YRD	Suburban	Summer				6.4(3.6-7.4)					10-1000	(Gao et al., 2009)	
2004	Xinken	PRD	Rural	Annual	26%	0.5-5.2		2.2-19.8					3-10000	(Liu et al., 2008)	
2005-2006	China Sea		Sea	Annual				3.4 / 3.5					15-10000	(Lin et al., 2007)	
2004-2005	Beijing	NCP	Urban	Annual	40%	3.3-81.4 / 22.3 / 16.2		0.1-11.2 / 1.8 / 2.4		2.5 / 6	0.59 / 2.4	0.014 / 0.031	3-10000	(Wu et al., 2007)	clean / pollution

*EP: events percentage (frequency).
**The average value and the range of the reported data in the parenthesis.