Evaluation of the contribution of new particle formation to cloud droplet number concentration in the urban atmosphere

Sihui Jiang1, Fang Zhang2, Jingye Ren1, Lu Chen1, Xing Yan1, Jieyao Liu1, Yele Sun3, and Zhanqing Li4

1College of Global Change and Earth System Science, Beijing Normal University, Beijing 100875, China
2Environmental Science and Engineering Research Center, School of Civil and Environmental Engineering, Harbin Institute of Technology (Shenzhen), 518055 Shenzhen, China
3State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China
4Earth System Science Interdisciplinary Center and Department of Atmospheric and Oceanic Science, University of Maryland, College Park, Maryland, USA

Correspondence: F. Zhang (fang.zhang@bnu.edu.cn)

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Abstract. The effect of new particle formation (NPF) on cloud condensation nuclei (CCN) varies widely in diverse environments. CCN or cloud droplets from NPF sources remain highly uncertain in the urban atmosphere; they are greatly affected by the high background aerosols and frequent local emissions. In this study, we quantified the effect of NPF on cloud droplet number concentration ($N_d$) at typical updraft velocities ($V$) in clouds based on field observations on 25 May–18 June 2017 in urban Beijing. We show that NPF increases the $N_d$ by 32 %–40 % at $V = 0.3–3$ m s$^{-1}$ during the studied period. The $N_d$ is reduced by 11.8 ± 5.0 % at $V = 3$ m s$^{-1}$ and 19.0 ± 4.5 % at $V = 0.3$ m s$^{-1}$ compared to that calculated from constant supersaturations due to the water vapor competition effect, which suppresses the cloud droplet formation by decreasing the environmental maximum supersaturation ($S_{\text{max}}$). The effect of water vapor competition becomes smaller at larger $V$ that can provide more sufficient water vapor. However, under extremely high aerosol particle number concentrations, the effect of water vapor competition becomes more pronounced. As a result, although a larger increase of CCN-sized particles by NPF events is derived on clean NPF days when the number concentration of preexisting background aerosol particles is very low, no large discrepancy is presented in the enhancement of $N_d$ by NPF between clean and polluted NPF days. We finally reveal a considerable impact of the primary sources on the evaluation of the contribution of NPF to CCN number concentration ($N_{\text{CCN}}$) and $N_d$ based on a case study. Our study highlights the importance of full consideration of both the environmental meteorological conditions and multiple sources (i.e., secondary and primary) to evaluate the effect of NPF on clouds and the associated climate effects in polluted regions.

1 Introduction

In the global climate system, aerosols, cloud condensation nuclei (CCN) and cloud droplets are very important components. Clouds, serving as a bridge connecting aerosols and climate, are the most uncertain factor of climate change (IPCC, 2013; Seinfeld et al., 2016; Cai et al., 2020). The microphysical link between aerosols and clouds is the most important part has received extensive attention. Cloud droplet activation is a key process from aerosol to clouds, and researchers have tried to simulate the microphysical processes using numerical activation models (e.g., Boucher and Lohmann, 1995; Abdul-Razzak et al., 1998; Ghan et al., 1993; Khvorostyanov and Curry, 1999; Abdul-Razzak and Ghan, 2000; Nenes et al., 2001, 2003; Petters and Kreidenweis, 2007; Ren et al., 2018; Genz et al., 2020).

New particle formation (NPF) events have been observed and occur frequently in different atmospheric environments in the world (Spracklen et al., 2010; Yue et al., 2011; Peng...
et al., 2017; Kerminen et al., 2018; Bousiotis et al., 2019; Zimmerman et al., 2020). NPF events are one of the most significant sources of fine particles in the atmosphere (Shi et al., 1999; Stanier et al., 2004; Kulmala and Kerminen, 2008). For example, it has been found that NPF contributed about 76% of the total fine particle number concentration in urban Beijing (Wu et al., 2011). These nucleated particles subsequently grow through coagulation or condensation processes to CCN-relevant sizes or act as CCN in convective clouds (Fan et al., 2013; Li et al., 2010). In reality, field studies have shown that these fine particles produced from NPF can subsequently result in an enhancement in N_{CCN} at cloud-relevant supersaturation (Kalkavouras et al., 2017; Peng et al., 2014; Wu et al., 2015; Ma et al., 2016; Z. Li et al., 2017; Zhang et al., 2019). It was estimated that up to 80% of CCN number concentration (N_{CCN}) is from the nucleation process in urban Beijing (Wiedensohler et al., 2008).

However, the N_{CCN} only reflects the cloud-forming potential of aerosol particles at a given supersaturation. The measurement of CCN is usually carried out at constant supersaturations. Different from the prescribed supersaturation used in the evaluation of N_{CCN}, when calculating the cloud droplet number concentration (CDNC, or N_{d}), researchers considered the dynamic situations in clouds. In clouds, the supersaturation exhibits variable levels that instantaneously adjust to the intensity of cloud updrafts and the particle number size distribution (PNSD) (Nenes et al., 2003; Hudson et al., 2015). So the CDNC (or N_{d}) depends on the size distribution, chemical properties of aerosol and the cloud updraft velocity, all of which regulate the maximum supersaturation (S_{max}) that can be formed in a cloud parcel (Nenes and Seinfeld, 2003). Studies have shown that the CDNC in clouds exhibits a sublinear relationship to aerosol number concentration (N_{CN}) (Twomey, 1977; Leaitch et al., 1986; Ghan et al., 1993; Boucher and Lohmann, 1995; Nenes et al., 2001; Ramanathan et al., 2001; Sullivan et al., 2016); this is different from CCN due to the limitation of the water vapor in the actual environment. Using the prescribed supersaturation to calculate CDNC may therefore provide a bias in the evaluation of the aerosol indirect effect. For example, Kalkavouras et al. (2017, 2019) reported an average 12% enhancement of CDNC during two consecutive NPF episodes in the eastern Mediterranean, which was significantly smaller than the enhancement of N_{CCN} (~ 87%) during the NPF events. Hence, it is critical to fully consider the background meteorological conditions (e.g., using dynamic water vapor under different updraft velocities) to simulate the S_{max} when evaluating the effect of NPF on clouds and the associated climate effects.

Relevant studies have been carried out in clean regions, but fewer were conducted in polluted urban areas. Field studies have shown that NPF events can occur frequently in polluted urban sites, although the high concentration of background particles is not conducive to the generation of new particles (Wu et al., 2011; Peng et al., 2014; Zimmerman et al., 2020), and the formation and growth rate of new particles may be larger than those of a relatively clean atmosphere. Wiedensohler et al. (2012) found that, under the high concentration levels of gaseous pollutants and strong oxidation in polluted areas, the high concentration of nanoparticles generated by NPF events can rapidly grow to tens or even hundreds of nanometers in a few hours. Zhang et al. (2019) observed that the subsequent growth of newly formed particles can last 2–3 d in urban Beijing, producing more CCN-sized particles. Previous studies in polluted regions demonstrated the complex and nonlinear relationship between aerosol particles and CCN due to multiple emission sources (Zhang et al., 2014, 2016, 2017, 2019; Ren et al., 2018; Fan et al., 2020), highlighting the importance of understanding the connections between aerosols and CCN or cloud droplets close to the source regions. Particularly, owing to the extremely high CN number concentrations (with order of magnitude as high as 10^{4} or even 10^{5} cm^{-3}) during NPF events in the urban area, the effect of competition for water vapor and reduction in cloud supersaturation is expected to be exacerbated.

The current study quantifies the contribution of NPF to N_{CCN} and CDNC in the polluted urban atmosphere of Beijing using field measurements of aerosol number size distributions and chemical composition. The effect of water vapor competition on evaluating N_{d} during NPF events is examined. The impact of the background preexisting particles on the enhancement of CCN and CDNC is also discussed by contrasting the results on typical clean NPF days and polluted NPF days. Given the strong local primary sources like traffic emissions in the urban area, a case study is conducted to investigate the impact of primary emissions on the evaluation of the effect of NPF on N_{d}.

2 Methodology
2.1 Site and experiment

A field campaign was conducted from 25 May to 18 June 2017 at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (39.98°N, 116.39°E), for measurements of aerosol physical and chemical properties. The IAP is located between the North Third Ring Road and Fourth Ring Road in northern Beijing. It is a typical urban background site, mainly affected by traffic and cooking emissions. Beijing is hot in summer, with high ambient relative humidity, which is conductive to the generation of atmospheric convection and reduces the high background aerosol condensation sink. The radiation in summer is stronger than other seasons, which promotes the generation of nucleated particles. In addition, local sources from traffic and cooking emissions, which may contribute many CCN-size-relevant particles, can be important at the site (Sun et al., 2015). The instruments during the campaign were deployed in a container at ground level (~8 m on a meteorological tower).
The number size distribution of particles in the size range from 10 to 550 nm (scanned range) was measured with a time resolution of 5 min by a scanning mobility particle sizer (SMPS; Wang and Flagan, 1990; Collins et al., 2002), which consists of a differential mobility analyzer (DMA, model 3081L, TSI Inc.) to classify particles with different sizes of particles, and a condensation particle counter (CPC, model 3772, TSI Inc.) to detect the size-classified particles. The sampled particles were dried to a relative humidity < 30% before entering the DMA. The non-refractory chemical composition of PM_{1} is measured by an aerosol chemical speciation monitor (ACSM), which consists of an aerodynamic lens to efficiently sample and focus submicron particles into the ACSM (Ng et al., 2011). Before sampling into the ACSM, aerosol particles are dried by silica gel desiccant. The ACSM was operated at a time resolution of 15 min. And the non-refractory chemical components that can be measured mainly include organics, sulfate salts (SO_{4}^{2-}), nitrate salts (NO_{3}^{-}), ammonium (NH_{4}^{+}) and chloride (Cl^{-}) (Ng et al., 2011). The refractory components mainly include black carbon (BC), and the BC mass concentration was measured using a seven-wavelength aethalometer (AE33, Magee Scientific Corp). 

### 2.2 Calculation of \( N_{CCN} \)

According to the hygroscopic growth process of particles described by Köhler theory (Köhler, 1936), the particles with a dry particle diameter (\( D_{p} \)) larger than the critical dry particle diameter (\( D_{c} \)) can be activated to form a cloud droplet. In this study, the \( \kappa \)-Köhler theory (Petters and Kreidenweis, 2007), which simply describes the approximate relationship between the \( D_{c} \) with the critical supersaturation (\( S_{c} \)), is applied as follows, when \( \kappa > 0.1 \):

\[
K = \frac{4A^{3}}{27D_{c}^{3} \ln S_{c}}, \quad A = \frac{4\sigma_{w}M_{w}}{RT \rho_{w}},
\]

where \( M_{w} \) is the molecular weight of water (\( M_{w} = 0.018015 \text{ kg mol}^{-1} \)), \( \rho_{w} \) is the density of water (\( \rho_{w} = 997.1 \text{ kg m}^{-3} \)), \( T \) is the parcel temperature (\( T = 298.15 \text{ K} \)), where \( \sigma_{w} \) is the droplet surface tension at the point of activation (\( \sigma_{w} = 0.072 \text{ J m}^{-2} \)), and \( R \) is the universal gas constant (\( R = 8.315 \text{ J K}^{-1} \text{ mol}^{-1} \)). \( \kappa \) is a hygroscopic parameter which depends on the chemical composition of the particle. In this study, based on the assumption that particles are internally mixed and their chemical composition will not be impacted by changes in particle size, we derived the \( \kappa \) with a simple mixing rule on the basis of chemical volume fractions (Petters and Kreidenweis, 2007; Gunthe et al., 2009). We used ACSM data, combined with the positive matrix factorization (PMF) analysis data to calculate the organic and inorganic volume fraction, according to the following equation:

\[
\kappa_{\text{chem}} = \sum_{i} \varepsilon_{i} \kappa_{i},
\]

where \( \kappa_{i} \) and \( \varepsilon_{i} \) are the hygroscopic parameter and volume fraction for each individual (dry) component in the mixture, respectively. The \( \kappa \) value and density (\( \rho \)) of each species used in the calculation are given in Table 1, which are from Petters and Kreidenweis (2007) and Topping (2005).

In Eq. (1), the corresponding \( D_{c} \) can be obtained from a given \( S_{c} \), and all particles with diameters larger than \( D_{c} \) can be activated. So the \( N_{CCN} \) can be calculated by integrating the PNSD from \( D_{c} \) to the largest particle size measured:

\[
CCN(D_{c}) = \int_{D_{c}}^{550} n(\log D_{p})d \log D_{p},
\]

where \( n(\log D_{p}) \) is the particle number that corresponds to each particle size bin \( d \log D_{p} \) in the aerosol number size distribution.

### 2.3 Calculation of \( N_{d} \)

The \( N_{d} \) depends on the \( S_{\text{max}} \) that can be formed in adiabatic ascending clouds. And this “cloud-relevant” supersaturation varies at different updraft velocity. A global scheme of cloud droplet parameterization has been established and developed for the calculation of the \( N_{d} \) and \( S_{\text{max}} \) (Nenes and Seinfeld, 2001; Fountoukis and Nenes, 2005). In this study, the \( S_{\text{max}} \) was calculated from an equation that expresses the water vapor balance in adiabatic ascending cloud (Nenes and Seinfeld, 2003):

\[
\frac{dS}{dt} = \alpha V - \gamma \frac{dw}{dr},
\]

where \( \alpha \) and \( \gamma \) are two coefficients that can be calculated by meteorological constants, the product of \( \alpha \) and \( V \) expresses the increase of supersaturation due to the adiabatic cooling of the parcel, and \( \frac{dw}{dr} \) denotes the water condensation rate during the aerosol activation and subsequent growth processes, shown in detail in Eq. (4). And \( \frac{dS}{dt} \) expresses the growth rate of supersaturation; when it is equal to 0, the supersaturation reaches the maximum value.

\[
\frac{dw}{dr} = \frac{\pi}{2} \omega \int_{0}^{S_{\text{max}}} D_{p}^{2} n(S') dS',
\]

where \( \omega \) is the density of water. \( n(S')dS' \) is the number concentration of particles activated between \( S' \) and \( S' + ds' \).

Nenes et al. (2001) used a sectional representation of the CCN spectrum (i.e., particle number supersaturation distribution \( n(S') \)) and total number of particles with \( S_{c} \) smaller than \( S_{c} \), \( F_{S}(S) \), which is given by

\[
F_{S}(S_{c}) = \int_{0}^{S_{c}} n(S') dS',
\]

where \( S_{c} \) is the supersaturation in the environment and \( n(S') \) represents the number concentration of particles activated between \( S' \) and \( S' + ds' \) in the CCN spectrum. The
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Table 1. Densities of different chemical species and their $\kappa$ measured by the laboratory.

| Species      | NH$_4$NO$_3$ | (NH$_4$)$_2$SO$_4$ | NH$_4$HSO$_4$ | H$_2$SO$_4$ | POA* | SOA* | BC   |
|--------------|--------------|-------------------|--------------|-------------|------|------|-----|
| $\rho$ (kg m$^{-3}$) | 1720         | 1769              | 1780         | 1830        | 1000 | 1400 | 1700 |
| $\kappa$    | 0.58         | 0.48              | 0.56         | 1.19        | 0    | 0.09 | 0    |

$\rho$ refers to primary organic aerosol, and SOA* refers to secondary organic aerosol.

2.4 Method for calculating the contribution of NPF to $N_{CCN}$ and $N_d$

The increment of $N_{CCN}$ or $N_d$ by the NPF ($\Delta N_{CCN}$ or $\Delta N_d$) is usually quantified by comparing the $N_{CCN}$ or $N_d$ prior to and after the NPF event (Peng et al., 2014; Wu et al., 2015; Ma et al., 2016; Ren et al., 2018; Zhang et al., 2019; Fan et al., 2020). In this study, the $N_{CCN}$ or $N_d$ prior to the NPF event was determined as a 2 h average of $N_{CCN}$ or $N_d$ before the burst of newly formed nucleated particles. And the $N_{CCN}$ and $N_d$ after the NPF event were calculated as the average of $N_{CCN}$ or $N_d$ from the beginning to the end of the NPF impact on the $N_{CCN}$ or $N_d$. So it is critical to determine when a NPF event starts and ends when NPF begins and ends its impact on the $N_{CCN}$ or $N_d$.

Generally, the burst in the nucleation-mode particles symbolizes the beginning of an NPF event. Here, the moment when a half-hour concentration of the nucleation-mode particles suddenly increases by orders of magnitude as high as $\sim 10^9$ cm$^{-3}$ during NPF cases was defined as $t_{\text{start}}$. The end time of an NPF event, $t_{\text{end}}$, is defined by the moment when the half-hour concentration of nucleated particle is lower than that at $t_{\text{start}}$.

Since some time is needed for the newly formed nucleated particles to grow to a sufficient size to act as CCN, the $N_{CCN}$ would not be enhanced as soon as new particles are generated. To determine the time that NPF begins and ends its impact on the $N_{CCN}$, denoted as $t_{\text{start,CCN}}$ and $t_{\text{end,CCN}}$ respectively, the time series of $N_{CCN}$ was firstly divided by the $N_{CCN}$ at $t_{\text{start}}$ at each prescribed supersaturation, to derive the normalized time series of $N_{CCN}$, denoted as $R_S$. The equation is written as follows:

$$R_S = \frac{CCN_S}{CCN_{S,\text{start}}},$$

where $S$ represents the supersaturation. Before the new particles reach a large enough size to impact $N_{CCN}$, the variations of $R_S$ should remain constant for different supersaturations if the concentrations of the background or preexisting aerosols change insignificantly. And at $t_{\text{start,CCN}}$ when NPF begins to impact the $N_{CCN}$, an apparent increase in $R_S$ is observed, taking the observation on 11 June as an example (Fig. 1a). Also, due to the heterogeneous composition and distinct CCN activity of the newly formed particles (Duan et al., 2018; Ren et al., 2018; Zhang et al., 2019; Tao et al., 2021), a parameter, $R_d$, which was calculated with the relative standard deviation of the $R_S$ of different supersaturations at a given time, is applied to fix the $t_{\text{start,CCN}}$ and $t_{\text{end,CCN}}$. Then the $t_{\text{start,CCN}}$ and $t_{\text{end,CCN}}$ correspond to the moments when the $R_d$ starts to increase and goes back to nearly zero (Fig. 1b) between the $t_{\text{start}}$ and $t_{\text{end}}$. The same method is used to determine the time that NPF begins and ends its impact on the $N_d$, denoted as $t_{\text{start},N_d}$ and $t_{\text{end},N_d}$ respectively (Fig. 1d, e).

More details about the method can be found in Kalkavouras et al. (2019). As shown in Fig. 1, it is clear that both the $N_{CCN}$ and $N_d$ exhibit a large increase in the NPF-impacted time zone between $t_{\text{start},CCN}$ and $t_{\text{end},CCN}$ (Fig. 1c) and between $t_{\text{start},N_d}$ and $t_{\text{end},N_d}$ (Fig. 1f). The average time lag between $t_{\text{start}}$ and $t_{\text{start},N_d}$ was about 3–5 h, which is shortened by 50 % compared to that reported by Kalkavouras et al. (2019). This case on 11 June was not a unique case, and similar patterns are also shown on other NPF days during the campaign (Figs. S3–S8).

Note that this method operates with an assumption of the unchanged background preexisting aerosols during the NPF events, without consideration of the impacts from local emission sources and diurnal changes in the planetary boundary layer (PBL). As shown in Fig. 2b, the time series of $N_{CN}$ presents a baseline which indicates that the concentrations of the background aerosols on each of the 7 typical NPF days do not vary much; the impact from the variation of background aerosol particles thus should be insignificant. The impact of...
Figure 1. The diurnal evolution of (a) the $R_S$ of $N_{CCN}$ at different supersaturations, (b) the relative dispersion of $R_S$ and $R_d$ for $N_{CCN}$ at different supersaturations, (c) the calculated $N_{CCN}$ under different supersaturations, (d) the $R_S$ of $N_d$ under updraft velocities from 0.3 to 3.0 m s$^{-1}$, (e) the $R_d$ for $N_d$ and (f) the calculated $N_d$ at updraft velocities of 0.3 and 2.1 m s$^{-1}$ on 11 June 2017.

PBL is expected to be small when the growth of the newly formed particles spans only a few hours. However, when the growth continues over a longer period to evening or at night, which may coincide with the period during which the PBL height changes from high to low (Kerminen et al., 2012; Altstädtler, et al., 2015; Z. Li et al., 2017), it will result in a larger $N_{CCN}$ and $N_d$, leading to an overestimation of the contribution of NPF to $N_{CCN}$ and $N_d$. A quantitative evaluation of such an impact is difficult due to the contemporary PBL data not being available. Therefore, here we only investigate the impact of local emissions on the evaluation of the effect of NPF on $N_d$ based on a case study.

3 Results and discussion

3.1 Time series of observed NPF events and calculated $N_{CCN}$ and $N_d$

During the observed periods from 25 May to 15 June 2017, the NPF events occurred on most days ($\sim$ 13 d) (Fig. 2a). According to Dal et al. (2005) and Wu et al. (2015), a typical NPF event includes the sudden appearance and continuous growth of particles smaller than 25 nm, and a “banana” shape can be seen on the particle number size spectrum. While non-NPF events may also have sudden increases of fine particles at a short timescale (e.g., local sources from vehicle or cooking emissions), they do not show a banana shape. Therefore, those cases with a typical banana shape (seven NPF events in total), which presents a complete NPF evolution process from nucleation to subsequent growth (not interrupted by meteorological conditions either), are selected for further study (marked by pink shading in Fig. 2). Figure 2b, c and d present the time series of $N_{CN}$, $N_{CCN}$ and $N_d$. The figure shows that the NPF event drives the variation of $N_{CCN}$ and $N_d$, confirming that the occurrence of NPF events is an important source of CCN. The variation trend of $N_{CCN}$ is more correlated with that of $N_{CCN}$ than $N_d$ (see also Fig. 4, Table S5). This is because the $N_{CCN}$ was calculated based on a constant $S$ rather than referring to the availability of water vapor, while the calculation of $N_d$ is based on the $S_{max}$ that can be reached in the real atmosphere at a given updraft velocity. In the cloud, the change in the quantity of cloud particles can be directly reflected by the change in $S_{max}$. As shown in Fig. 2e, the average $S_{max}$ for the two vertical updraft velocity of $V = 0.3$ and $V = 2.1$ m s$^{-1}$ was calculated to be under 0.2 % and 0.4 %, varying largely with the variation of $N_{CN}$ due to the effect of water vapor competition, which will be discussed in Sect. 3.3.

3.2 Quantitative evaluation of the NPF impact on $N_{CCN}$ and $N_d$

Based on the method in Sect. 2.4, the contribution of the NPF to $N_{CCN}$ and $N_d$ is calculated and is shown in Fig. 3. The results show that the $N_{CCN}$ is increased on average by 32.0 %, 43.0 %, 53.0 % and 65.0 % at $S$ of 0.2 %, 0.4 %, 0.6 % and 0.8 % respectively during NPF events (Fig. 3b, c, Table S3), amounting to about 24 %–37 % of environment CCN at the cloud-relevant supersaturation being directly originated from NPF during the studied period in urban Beijing. And the rest (about 63 %–76 %) of CCN are from other sources or preexisting particles, which is a much larger proportion than that derived in remote Finokalia, Crete, Greece, by Kalkavouras et al (2019). In other words, due to the higher background concentration of aerosol particles in the polluted urban area,
the relative contribution of NPF to \( N_{\text{CCN}} \) is more significant in remote clean regions.

The estimated results of \( N_d \) for selected vertical updraft velocities are shown in Fig. 3d–f. Generally, the average \( S_{\text{max}} \) was calculated to be under 0.4\% and 0.2\% for \( V = 2.1 \) and \( V = 0.3 \) m s\(^{-1}\) respectively (Fig. 2e), corresponding to critical particle sizes \( (D_c) \) of \( \sim 70 \) and \( \sim 110 \) nm. This means that most activated drops are from accumulation-mode particles and larger particles in Aitken mode. The large contribution of the Aitken-mode particles leads to a large number of cloud droplets in urban Beijing, especially for high updraft velocity. Basically, the average \( N_d \) values (increased by NPF) are 433, 854, 1117, 1281 and 1523 cm\(^{-3}\) at updraft velocities of 0.3, 0.9, 1.5, 2.1 and 3 m s\(^{-1}\) respectively (Fig. 3d), which is a much larger magnitude compared with that in the clean areas (Morales Betancourt et al., 2014; Sullivan et al., 2016; Kalkavouras et al., 2019). This is equal to the \( N_d \) being enhanced by 32\%, 37\%, 38\%, 38\% and 40\% at updraft velocities of 0.3, 0.9, 1.5, 2.1 and 3 m s\(^{-1}\) respectively (Fig. 3e), suggesting that the higher cloud updraft velocity not only generates more cloud droplets, but also induces larger enhancements in \( N_d \). We also show that the NPF contributes about 30\% to the total \( N_d \) during the studied period in urban Beijing (Fig. 3f). And the rest (about 70\%) of cloud droplets are from the other sources or preexisting particles. With the increase of the \( S \), the percentages of NPF-initiated \( N_{\text{CCN}} \) and the contributions of the NPF to \( N_{\text{CCN}} \) increased more significantly than for \( N_d \) with the increase of \( V \). In other words, the percentages of NPF-initiated \( N_d \) and the contributions of the NPF to \( N_d \) are relatively independent of the variation of \( V \). This is primarily due to the water vapor competition effect under very high CN number concentrations when calculating the \( N_d \). Under high \( N_{\text{CN}} \), the water vapor competition effect will lead to lower \( S_{\text{max}} \), which is smaller than the constant \( S \) for calculating \( N_{\text{CCN}} \). Roughly, the \( N_d \) at \( V = 0.3–3 \) m s\(^{-1}\) corresponds to the \( N_{\text{CCN}} \) at \( S = 0.1–0.5 \)%, within which the percentages of \( \Delta N_{\text{CCN}} \) and the contributions of the NPF to \( N_{\text{CCN}} \) do not change much either. The effect of water vapor competition will be further examined in the following section.

### 3.3 The effect of water vapor competition on evaluating \( N_d \)

Figure 4 shows the scatter plots of correlations between \( N_{\text{CN}} \) and \( N_{\text{CCN}} \) at supersaturations of 0.6\% and 0.8\% and the correlations between \( N_{\text{CN}} \) and \( N_d \) under updraft vertical velocities of 2.1 and 3.0 m s\(^{-1}\). The \( N_{\text{CCN}} \) and \( N_{\text{CN}} \) were obviously linearly related, but the correlation between \( N_d \) and \( N_{\text{CN}} \) was nonlinear. When shown as average values with error bars, the \( N_d \) increases linearly as \( N_{\text{CN}} \) increases when the \( N_{\text{CN}} \) is below 15 000, then the \( N_d \) begins to decrease with the further increase of \( N_{\text{CN}} \). This has been presented in previous studies (Nenes et al., 2001; Ramanathan et al., 2001; Sullivan et al., 2016) and is believed to be caused by the water vapor competition of the aerosol particles. Although the larger updraft velocities can achieve greater supersaturation in adiabatic ascending clouds, and more particles can be activated into cloud droplets, the water vapor competition still occurred when background aerosol particles increased...
Figure 3. Box diagram of the increment of (a) CCN number concentration $N_{CCN}$ ($\Delta N_{CCN}$), (b) enhanced percentage of $N_{CCN}$ and (e) the contribution to total $N_{CCN}$ by NPF for different supersaturations (0.2 %–0.8 %) and (d) cloud droplet number concentration $N_d$ ($\Delta N_d$), (e) enhanced percentage of $N_d$ and (f) the contribution to total $N_d$ by NPF under different updraft velocities (0.3–3.0 m s$^{-1}$).

to a certain number. This is strongly suggested by the difference between the calculated $N_{CCN}$ using the constant $S$ and the $N_d$ using the variable $S_{\text{max}}$ in the air parcels because in the actual environment, it is often not possible to achieve sufficient supersaturation compared to the prescribed levels that are preset by the instrument. For example, the average $S_{\text{max}}$ is lower than 0.5 % at the maximum cloud updraft velocity of 3 m s$^{-1}$ according to the calculation results in this study. Therefore, although NPF events may strongly increase $N_{CCN}$, the formed $N_d$ is eventually limited by water vapor competition, which determines the $S_{\text{max}}$ that varies in the cloud. The $S_{\text{max}}$ is related to the cloud formation dynamics and the aerosol levels in the region.

To evaluate the effect of water vapor competition on $N_d$, by taking the case on 11 June as an example, we compare the $N_d$ calculated from the varied $S_{\text{max}}$ at different updraft velocities with the $N_d$ at constant $S$ (Fig. 5). The results from other NPF cases are also summarized in Tables S6 and S7. Obviously, after the $I_{\text{start}}$, the $S_{\text{max}}$ starts to decrease and was negatively correlated with $N_d$ for both the updraft velocities, reflecting the enhanced effect of competition for water vapor from the growing number of droplets (Fig. 5a and b). It is shown that $S_{\text{max}}$ was decreased by $14.5 \pm 3.5 \%$, $13.3 \pm 4.0 \%$, $13.4 \pm 4.2 \%$, $12.0 \pm 4.1 \%$ and $11.7 \pm 3.9 \%$ for $V = 0.3, 0.9, 1.5, 2.1$ and $3$ m s$^{-1}$ respectively (Fig. 5c, d).

Therefore, in comparison to the $N_d$ calculated from the constant $S$, the $N_d$ calculated from the variable $S_{\text{max}}$ is greatly reduced at both the updraft velocities of 0.3 m s$^{-1}$ and 2.1 m s$^{-1}$, suggesting a significant suppression of cloud droplet formation. Quantitatively, the $N_d$ is reduced by $19.0 \pm 4.5 \%$, $15.7 \pm 4.7 \%$, $14.8 \pm 5.6 \%$, $12.3 \pm 4.9 \%$ and $11.8 \pm 5.0 \%$ at updraft velocity of 0.3, 0.9, 1.5, 2.1 and 3 m s$^{-1}$ respectively on the NPF days. Our results are similar to those reported by Kalkavouras et al. (2017), who showed that these competition effects suppress $N_d$ by 20 % for $V = 0.3$ and 12.3 % for $V = 0.6$ m s$^{-1}$. In addition, the declined percentages with an increase in updraft velocity suggest that the effect becomes smaller at larger $V$, achieving greater $S_{\text{max}}$ in the environment. Essentially, water vapor competition led to the reduction in $N_d$ by decreasing the required $S_{\text{max}}$ for the CN activation.

### 3.4 The variations of CCN and cloud droplet on typical clean and polluted NPF days: a case study

Generally, lower PM$_{2.5}$ means a low background condensation sink (CS), which is conducive to the condensation and coagulation of nucleation particles (Wu et al., 2011; Yue et al., 2011; Wiedensohler et al., 2012). Different from the remote clean area, some of the NPF events in urban Beijing during the campaign occurred with background pollution (with daily mass concentrations of PM$_{2.5}$ of ~ 40 $\mu$g m$^{-3}$) or are impacted by local primary emissions. This kind of NPF event has different characteristics from that in clean conditions, as the sudden increase of nucleation particles less than 25 nm is often accompanied by an increase of large particles at the beginning of NPF. Here, they are named as polluted and clean NPF events respectively. Two days, on 27 May and 11 June, representing the typical polluted and clean NPF events respectively. Two days, on 27 May and 11 June, representing the typical polluted and clean NPF events respectively, are selected for contrasting the effect of the two kinds of NPF on CCN and CDNC. As shown in Fig. 7, there is a higher preexisting background of accumulation-mode particles throughout the day on the polluted NPF day of 27 May than on the clean NPF day of 11 June. On the clean NPF days, many more nucleation- and Aitken-mode particles, with $N_{CCN}$ enhancement 2-fold higher than that on polluted days (Fig. 7a), were generated, and NPF
events developed more strongly in the initial stage. The beginning of NPF events ($t_{\text{start}}$) in the polluted case (11:00) was about 2 h later than that in the clean case ($\sim$ 09:00).

For both cases, the $N_{\text{CCN}}$ is increased with the evolution of the NPF events (Fig. 6a, b, e, f). But the magnitude of the enhancements in the two cases is quite different. The $N_{\text{CCN}}$ during NPF events on polluted days was generally twice than that of clean days (Fig. 6e and f) because there were a large number of preexisting CCN-sized aerosol particles on polluted NPF days. As a result, a larger increment of $N_{\text{CCN}}$ is derived on clean NPF days, showing 37%–80% and 25%–41% percentage increases of $N_{\text{CCN}}$ from NPF on clean and polluted days respectively (Fig. 7b). For $N_d$, the percentage increase on clean days was 22% and 37% and on polluted days 34% and 26% under updraft velocities of 0.3 and 2.1 m s$^{-1}$. The percentage increases in $N_d$ between clean and polluted days are comparable. This result further illustrates the effect of water vapor competition on $N_d$ under high $N_{\text{CN}}$ in the polluted atmosphere. This suggests that it is critical to fully consider the background meteorological conditions (e.g., using dynamic water vapor under different updraft velocities) to simulate the $N_d$ when evaluating the effect of NPF on clouds and the associated climate effects.

Figure 4. Scatter plots of correlation between total number concentration ($N_{\text{CN}}$) and CCN number concentration ($N_{\text{CCN}}$) at supersaturation of (a) 0.6% and (c) 0.8% respectively. Scatter plot of correlation between $N_{\text{CN}}$ and cloud droplet number concentration ($N_d$) at updraft vertical velocities of (b) 2.1 m s$^{-1}$ and (d) 3.0 m s$^{-1}$ respectively.

Figure 5. (a) The diurnal changes of the calculated maximum supersaturation ($S_{\text{max}}$) (referring to the constant supersaturation $S$) at $V = 0.3$ and 2.1 m s$^{-1}$ on 11 June 2017. (b) Comparison of the cloud droplet number concentration ($N_d$) under the constant $S$ and $S_{\text{max}}$ on 11 June 2017. The suppression percentage of (c) $S_{\text{max}}$ and (d) $N_d$ due to the competition of water vapor.
Figure 6. Comparison of (a, b) the particle number size distribution PNSD, (c, d) aerosol particle number concentration $N_{CN}$, (e, f) CCN number concentration $N_{CCN}$ and (g, h) cloud droplet number concentration $N_d$ between a clean and a polluted NPF event. The clean NPF day is with a clean background ($PM_{2.5} = 14$ (µg/m$^3$)), and the polluted NPF day is with $PM_{2.5}$ of 73 µg/m$^3$.

Figure 7. Comparison of the increments of (a) total particle number concentration ($N_{CN}$) and (b) CCN number concentration ($N_{CCN}$) and cloud droplet number concentration ($N_d$) between the two different typical NPF events.
Table 2. Quantitative evaluation of the contribution of primary emissions to \( N_d \) and \( N_{CCN} \).

| \( V \) or \( S \) (m s\(^{-1}\) or %) | \( D_c \) (nm) | \( N_{d_{NPF}} \) or \( \Delta N_{CCN,NPF} \) (cm\(^{-3}\)) | \( N_{d_{PE}} \) or \( \Delta N_{CCN,PF} \) (cm\(^{-3}\)) | \( N_{d_{total}} \) or \( N_{CCN,total} \) (cm\(^{-3}\)) |
|----------------|---------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|
| Evaluation of the contribution of primary emissions to \( N_d \) |
| 0.3            | 140           | 200                                          | 84.4 %                                        | 37                                            |
| 0.9            | 107           | 543                                          | 86.6 %                                        | 84                                            |
| 1.5            | 93            | 676                                          | 87.5 %                                        | 97                                            |
| 2.1            | 84            | 750                                          | 83.1 %                                        | 153                                           |
| 3.0            | 75            | 942                                          | 77.1 %                                        | 279                                           |

| Evaluation of the contribution of primary emissions to \( N_{CCN} \) |
|----------------|---------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|
| 0.2 %          | 109           | 654                                          | 92.0 %                                        | 57                                            |
| 0.4 %          | 69            | 1356                                         | 87.2 %                                        | 199                                           |
| 0.6 %          | 52            | 1680                                         | 87.1 %                                        | 249                                           |
| 0.8 %          | 43            | 1801                                         | 85.0 %                                        | 318                                           |

\(^a\) PE, primary emission.

3.5 The impact of primary emissions during evening rush hour on the calculation of the contribution of NPF to \( N_{CCN} \) and \( N_d \): a case study

During the campaign, very high number concentrations of fine particles were observed during evening rush hour (as shown in Fig. 2a) when primary emissions related to automobile exhaust or cooking activities near the site may impact the PNSD. Those particles from primary emissions can serve as CCN and thereby impact the evaluation of the contribution of NPF to \( N_d \). Therefore, taking the day of 11 June as an example, such an effect from primary emissions during evening rush hour is investigated (Fig. 8). On the day, 1 h after the burst of newly formed particles at \( \sim 12:00 \), the \( N_d \) began to rise rapidly, and the increase of \( N_d \) continued until 21:30 (Fig. 1f). At \( \sim 18:00 \), the primary emissions also begin to impact the \( N_{CCN} \) and \( N_d \). Note that a sudden decrease and dilution in the PNSD are due to a precipitation event at \( \sim 21:30 \). From 18:00–21:30, the CCN and cloud droplets were from both the NPF source and the primary emissions. The impact of primary emissions is also indicated by the variations of particles composition during 18:00–22:00, when both the primary organic aerosols (POAs) and BC show a rapid increase in the mass concentration and fraction (Fig. 8c and d). Here, a positive matrix factorization (PMF) analysis was performed to separate the primary and secondary organic aerosol factors quantitatively for the purpose of source apportionment based on field measurement by an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) (Xu et al., 2017; Zhang et al., 2011). The PMF algorithm in the robust mode (Paatero and Tapper, 1994) was applied to the high-resolution mass spectra to resolve distinct organic aerosol factors representing primary and secondary sources and processes. More details about operation of the HR-ToF-AMS and PMF analysis also can be found in the supporting information of Liu et al. (2021).

To evaluate the impact of the primary emissions, it is critical to separate the particle modes representing the primary aerosols from the observed PNSD. According to the observed characteristics of PNSD, the newly formed particles continue to grow and are dominated by Aitken mode for several hours after the NPF occurred (Fig. 8a). The size mode of the newly formed particles during the rush hour is estimated by applying a growth rate of \( 3.2 \pm 0.5 \) nm h\(^{-1}\), which is calculated by the variation of median particle size during 12:00–18:00. The calculation results show that the NPF-tracked particles can grow to \( \sim 50–60 \) nm during the rush hour period. The primary particles from vehicles or cooking generally have a smaller size (\( \sim 30 \) nm) than the NPF-tracked mode and accumulation mode (\( \sim 100–120 \) nm) (Brines et al., 2015; Dall’Osto et al., 2011; Harrison et al., 2011), so we applied three modes to fit the PNSD from the beginning of the evening rush hour to the end, assuming a normal distribution. Note that the size mode for background aerosols almost coincides with the accumulation mode of primary emitted particles during the period. Since the mode and concentration of background aerosols do not change much before and after the occurrence of new particles (Fig. 8a, b), the impact of background aerosol is thus deducted from the fitting accumulation mode. The fitted result shows a major peak in the Aitken mode at \( \sim 50 \) nm that is related to the NPF event and two minor peaks in Aitken (\( \sim 30 \) nm) and accumulation (\( \sim 100–120 \) nm) mode (Fig. 8e, f) that are associated with the primary vehicle or cooking emissions. Figure 8g and h show the separated PNSD of the NPF-related and primary aerosols respectively. Then the increments of \( N_{CCN} \) and \( N_d \) from NPF are obtained from the PNSD of NPF mode, and the increments of \( N_{CCN} \) and \( N_d \) from primary emissions are
Figure 8. Diurnal variations of the (a) aerosol size distribution, (b) particle number concentrations for different size modes, (c) mass concentrations of aerosol chemical composition and (d) mass fraction of aerosol chemical components, (e, f) fitted three modes of the particle number concentration PNSD at 18:00 and 21:30, and (g) diurnal variations of the separated NPF-related PNSD and (h) the PNSD of primary aerosols.

obtained by subtracting the increment of $N_{CCN}$ and $N_d$ by NPF from the total increment of $N_d$.

The calculated results are summarized in Table 2. For $N_d$, the average contribution of primary emission to $N_d$ is 15.6 %, 13.4 %, 12.5 %, 16.9 % and 22.9 % cm$^{-3}$ for updraft velocities of 0.3, 0.9, 1.5, 2.1 and 3 m s$^{-1}$ respectively. The proportion of contribution from NPF and primary emission to the $N_d$ increment changes with the variation of $V$. The higher proportion of contribution from primary emission is obtained at higher $V$, which may be determined by the different characteristics between atmospheric particles emitted from the evening traffic sources and generated from NPF events. For $N_{CCN}$, the average contribution from primary emissions is 8.0 %, 12.8 %, 12.9 % and 15.0 % at $S$ of 0.2 %, 0.4 %, 0.6 % and 0.8 % respectively. Compared with $N_d$, the contribution percentage of primary emission to $N_{CCN}$ is smaller due to the total $N_{CCN}$ being much more than the total $N_d$. Our results show a considerable impact of these primary sources when evaluating the contribution of NPF to cloud droplet number, highlighting the importance of considering the influence from multiple (i.e., secondary and primary) sources on clouds in the polluted atmosphere. Finally, it is worth noting that the dynamic changes of PBL would also impact the $N_{CCN}$ and $N_d$ during the period, and the decrease in the height of PBL from the daytime to evening will result in an increase of $N_{CCN}$ or $N_d$. However, for this case, the impact from primary emissions is much more prominent, as indi-
cated by the sharply raised particle number concentrations during the rush hour (Fig. 8b).

4 Conclusions

In this study, we quantified the contribution of NPF to $N_d$ at typical updraft velocities in clouds using field measurements of aerosol number size distributions and chemical composition in urban Beijing. We show that NPF drives the variations of $N_{CCN}$ and $N_d$. About 32%–65% $N_{CCN}$ are increased by NPF events for supersaturation of 0.2%–0.8% in the polluted atmosphere. And the $N_d$ is increased about 32%–40% by NPF at $V = 0.3–3$ m s$^{-1}$ accordingly. A significant reduction in $N_d$ is observed due to water vapor competition with consideration of actual environmental updraft velocity, with decrease rates of 11.8% ± 5.0% at $V = 3$ m s$^{-1}$ and 19.0% ± 4.5% at $V = 0.3$ m s$^{-1}$ in comparison with a constant supersaturation. The effect of water vapor competition becomes smaller at larger $V$, at which a greater $S_{\text{max}}$ can be achieved. Essentially, water vapor competition led to the reduction in $N_d$ by decreasing the environmental $S_{\text{max}}$ for the activation of aerosol particles. It is shown that $S_{\text{max}}$ was decreased by 14.5 ± 3.5% to 11.7 ± 3.9% for $V = 0.3–3$ m s$^{-1}$. Our results suggest significant suppression of cloud droplet formation due to the water vapor competition, particularly at extremely high aerosol number particle concentrations. As a result, although a larger enhancement of CCN-sized particles by NPF event is derived on clean NPF days when there are few preexisting background aerosol particles, there is no large discrepancy in the enhancement of $N_d$ by NPF between the clean and polluted NPF days. Finally, we show a considerable impact of the primary sources when evaluating the contribution of NPF to cloud droplet number using a case study. Our study highlights the importance of full consideration of both the environmental meteorological conditions and multiple sources (i.e., secondary and primary) to evaluate the effect of NPF on clouds and the associated climate effects. For example, Merikanto et al. (2010) used model to simulate the variation of CDNC from the year 1850 to 2000 and showed that NPF made a nearly equal contribution (16%–13.5%) to global CDNC in all those years, leading to about a 50% enhancement in the year from 1850 to 2000 in cloud albedo. There are still large uncertainties in how to accurately quantitatively assess the response of these climate effects to NPF. This study is carried out in a polluted urban area and is beneficial for the research on aerosol–cloud microphysical processes. It provides a new perspective for follow-up research in the urban atmosphere. Note that there are still limitations of our studies, as we only investigated several NPF cases within a short period due to the limited measurement data. The small sample size might cause bias in the results. Further studies based on more measurement data, i.e., with longer time periods and more observational sites, are warranted to verify and refine our results, so as to parameterize the impact of NPF events on cloud, precipitation, and radiative forcing in models.

Data availability. All data used in the study are available at https://data.mendeley.com/datasets/hkkzbn4zv3/1 (last access: 4 January 2021, Zhang, 2021) or from the corresponding author upon request (fang.zhang@bnu.edu.cn).

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Competing interests. The authors declare that they have no conflict of interest.

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