RESPONSES TO REVIEWERS’ COMMENTS

Review#1

The authors used different time slots to calculate the enhancement of CCN and CDNC due to NPF, which is very confusing and makes the two sets of results incomparable. For CCN, the NPF period is defined as 12:00 to 16:00, though the definition of the pre-NPF period is not clear. For CDNC, however, the NPF period is defined as the period between tNd and tend, which corresponds to a very long time of about 10 hours. Given the big difference, the enhancement of CCN and CDNC has very different physical meanings. Besides, when calculating the enhancement of CCN, the author noted that “the end time is taken as 16:00 to avoid the interference of evening traffic emissions”. However, the time period used to calculate the CDNC enhancement essentially covers the entire evening rush hour. Isn’t there the same concern about the interference of traffic emissions? Also, Figure 2b shows that CCN peaks at about 21:00. Is this peak attributable to or partly attributable to NPF? If yes, this should be considered in the calculation of CCN enhancement since this period is indeed included in the calculation of CDNC enhancement. If not, what is the reason for this strong peak? In summary, I think the authors should use consistent and reasonable methods to estimate the two enhancement ratios.

Re: Thank you for your comments. This is good and key point for the evaluation of the NPF effect on CCN and cloud droplet. Therefore, during the revision, we have considered this issue carefully, and updated the CCN results using the updated evaluation methods. For the comparison, we also used the method to evaluate the cloud droplet to calculate the CCN results. In the revised version, more details of the method for calculating the contribution of NPF to $N_{CCN}$ and $N_d$ have been given (see Section 2.4). In addition, besides the cloud droplet, we also evaluated the impact of evening traffic emissions on $N_{CCN}$ as the reviewer suggested, see lines 450-465. The statements have been included in the revised text, see lines 200-235, or as follows, Lines 200-235:

“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 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 the NPF event was determined as two-hours 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 was calculated as the average of $N_{CCN}$ or $N_d$ from begin to the end of the NPF impact the $N_{CCN}$ or $N_d$. So it is critical to determine when a NPF event start and end, or when a NPF begins and ends the 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 with order of magnitude as high as $\sim 10^4$ cm$^{-3}$ during NPF cases was defined as $t_{start}$. The end time of an NPF event, $t_{end}$, is
defined by the moment when the half-hour concentrations of nucleated particle is lower than that at \( t_{\text{start}} \).

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

\[
R_s = \frac{CCN_s}{CCN_{s,\text{start}}}
\]

(8)

where \( S \) represents the supersaturation. Before the new particles reaches a large enough size to impact \( N_{\text{CCN}} \), the variations of \( R_s \) should remain constant for different supersaturations if the concentrations of the background or pre-exist aerosols changes insignificant. And at \( t_{\text{start,CCN}} \) when NPF begin to impact the \( N_{\text{CCN}} \), an apparent increase in \( R_s \) is observed by taking the observation on June 11 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 back to nearly zero (Fig. 1b) respectively between the \( t_{\text{start}} \) and \( t_{\text{end}} \). The same method is used to determine the time that NPF begins and ends the impact on the \( N_d \), which are denoted as \( t_{\text{start,Nd}} \) and \( t_{\text{end,Nd}} \) respectively (Fig. 1d, e).

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

Lines 450-465:

“...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% \( \text{cm}^3 \) for updraft velocities of 0.3, 0.9, 1.5, 2.1 and 3 \( \text{m s}^{-1} \) respectively. The proportion of contribution from NPF and primary emission to \( N_d \) increment change 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 \( N_{\text{CCN}} \), the average contribution from primary emissions is 8.0%, 12.8%, 12.9%, 15.0% at \( S \) of 0.2%, 0.4%, 0.6%, 0.8% respectively. Compared with \( N_d \), the contribution percentage of primary emission to \( N_{\text{CCN}} \) is smaller due to that the total
$N_{CCN}$ is much more than the total $N_d$. Our result shows considerable impact of those primary sources when evaluating the NPF contribution 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 indicated by the sharply raised particle number concentrations during the rush hour (Fig. 8b).


### Table 2. Quantitative evaluation of the contribution of primary emissions to $N_d$ and $N_{CCN}$

<table>
<thead>
<tr>
<th>V or S</th>
<th>$D_i$</th>
<th>$\Delta N_{d,\text{NPF}}$ or $\Delta N_{CCN,\text{NPF}}$</th>
<th>$\Delta N_{d,\text{PF}}$ or $\Delta N_{CCN,\text{PF}}$</th>
<th>$\Delta N_{d,\text{total}}$ or $\Delta N_{CCN,\text{total}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>m s$^{-1}$ or %</td>
<td>nm</td>
<td>cm$^{-1}$</td>
<td>%</td>
<td>cm$^{-1}$</td>
</tr>
</tbody>
</table>

| Evaluation of the contribution of primary emissions to $N_d$ | |
|---|---|---|---|---|---|
| 0.3 | 140 | 200 | 84.4% | 37 | 15.6% | 237 |
| 0.9 | 107 | 543 | 86.6% | 84 | 13.4% | 627 |
| 1.5 | 93 | 676 | 87.5% | 97 | 12.5% | 773 |
| 2.1 | 84 | 750 | 83.1% | 153 | 16.9% | 903 |
| 3 | 75 | 942 | 77.1% | 279 | 22.9% | 1221 |
Evaluation of the contribution of primary emissions to $N_{CCN}$

<table>
<thead>
<tr>
<th>Emission</th>
<th>$D_0$ (nm)</th>
<th>CCN Efficiency</th>
<th>CDNC Efficiency</th>
<th>Pre-NPF</th>
<th>NPF period</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2%</td>
<td>109</td>
<td>654</td>
<td>92.0%</td>
<td>57</td>
<td>8.0%</td>
</tr>
<tr>
<td>0.4%</td>
<td>69</td>
<td>1356</td>
<td>87.2%</td>
<td>199</td>
<td>12.8%</td>
</tr>
<tr>
<td>0.60%</td>
<td>52</td>
<td>1680</td>
<td>87.10%</td>
<td>249</td>
<td>12.90%</td>
</tr>
<tr>
<td>0.80%</td>
<td>43</td>
<td>1801</td>
<td>85.00%</td>
<td>318</td>
<td>15.00%</td>
</tr>
</tbody>
</table>

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.

The enhancement ratios of CCN and CDNC due to NPF are both calculated using the increment during the NPF period relative to a pre-NPF period. Is this increment merely caused by NPF? In other words, even without NPF, are there other confounding factors that lead to the difference between these two periods? The
authors need to rule out the potential impact of other confounding factors.

Re: Yes, there are other potential factors affecting the enhancement of $N_{CCN}$ and CDNC during this period, such as primary emission, variation of background conditions the development of the boundary layer. The relevant statements and discussions have been included in the revised text, see lines 241-252 and 389-393, or see as follows,

Lines 241-252 “…Note that this method is with an assumption of the unchanged background pre-exist 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 concentrations of the background aerosols on each of the 7 typical NPF day don’t vary much, the impact from the variation of background aerosol particles thus should be insignificant. The impact of PBL is expected to be small when the growth of the newly formed particles spans only a few hours. However, when the growth continues longer time to evening or at night which may coincide with the period that the PBL height changes from high to low (Kerminen et al., 2012; Alstädter, et al., 2015; 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 impact is difficult due to that the contemporary PBL data is not available. Therefore, here we only investigate the impact of local emissions on the evaluation of NPF effect on $N_d$ based on a case study…”

Lines 389-393:

“…The result just further illustrates that the effect of water vapor competition on $N_d$ under high $N_{CN}$ in 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…”


Section 2.3: This section needs to be carefully revised. The meanings of many variables in the equations are never defined. Line 153, the term “population splitting”
is not understandable unless the readers are familiar with that literature. Line 158, Line 168, I believe equation (7) and equation (8) refer to wrong equations. Equation (5), “i” only appears on the right side but not on the left side. Line 164 and Line 166, the uppercase S and lowercase s are mixed up.

Re: Thanks a lot the careful check and comments. Some descriptions of the calculation method of the effect of NPF on CCN and cloud droplet are confusing. In the revision, we have rewritten and reorganized the section for introduce the method applied for calculation of the NPF contribution to both CCN and cloud droplet. We have also made careful corrections, including adding more words to explain the variables in the equations, unifying the symbols of “S”, etc. We believe the method has been clearly addressed after the major revision. The details of the method are given in Section 2.3 and 2.4, or see lines 182-188 and 201-235 or as follows:

Lines 182-188:

“…Nenes et al. (2002) 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 \(S\), \(F^S(S)\), which is given by

\[
F^S(S_c) = \int_0^{S_c} n^S(s')ds'
\]  

(6)

Where the \(S_c\) is the supersaturation in the environment, the \(n^S(s')\) in equation (6) represents the number concentration of particles activated between \(s'\) and \(s'+ds'\) in CCN spectrum. The \(F^S(S_c)\) can be calculated by the integration of \(n^S(s')\) from the lower limit 0 to upper limit \(S_c\)….”

Lines 201-235:

“…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 \((\Delta N_{CCN} \text{ or } \Delta N_d)\) is usually quantified by comparing the \(N_{CCN}\) or \(N_d\) prior 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 the NPF event was determined as two-hours 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 was calculated as the average of \(N_{CCN}\) or \(N_d\) from begin to the end of the NPF impact the \(N_{CCN}\) or \(N_d\). So it is critical to determine when a NPF event start and end, or when a NPF begins and ends the 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 with order of magnitude as high as \(10^4\) 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 concentrations of nucleated particle is lower than that at \(t_{\text{start}}\).

Since there needs some time for the newly formed nucleated particles to grow to 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 end the 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,
\[ R_S = \frac{CCN_S}{CCN_{S,start}} \] (8)

where \( S \) represents the supersaturation. Before the new particles reaches 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 pre-exist aerosols changes insignificant. And at \( t_{start,CCN} \) when NPF begin to impact the \( N_{CCN} \), an apparent increase in \( R_S \) is observed by taking the observation on June 11 as an example (Fig. 1a). Also, due to the heterogenous 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 \( t_{start,CCN} \) and \( t_{end,CCN} \). Then the \( t_{start,CCN} \) and \( t_{end,CCN} \) correspond to the moments when the \( R_D \) starts to increase and back to nearly zero (Fig. 1b) respectively between the \( t_{start} \) and \( t_{end} \). The same method is used to determine the time that NPF begins and ends the impact on the \( N_d \), which are denoted as \( t_{start,N_d} \) and \( t_{end,N_d} \) respectively (Fig. 1d, e).

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


A large part of the abstract (e.g., Line 19-29) is very difficult, if not impossible, to understand before finishing reading the entire manuscript. This part needs to be substantially rewritten to make it comprehensible without referring to the main text. 

Re: The abstract has been rewritten and revised, please see lines 24-41, or as follows,

“The new particle formation (NPF) effect on cloud condensation nuclei (CCN) varies widely in diverse environment. The CCN or cloud droplet from NPF sources remains highly uncertain in urban atmosphere which are greatly affected by the high background aerosols and frequent local emissions. In this study, we quantified the NPF effect on cloud droplet number concentration (CDNC, or \(N_d\)) at typical updraft velocities \((V)\) in clouds based on field observations on May 25-June 18, 2017 in urban Beijing. We show that the 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 suppress the cloud droplet formation by decreasing the environmental maximum supersaturation (\(S_{\text{env}}\)). 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-size particles by NPF event is derived on clean NPF day when the number concentration of pre-existing background aerosol particles is very low, no large discrepancy is presented in the enhancement of \(N_d\) by NPF between the clean and polluted NPF day. We finally reveal a considerable impact of the primary sources on the evaluation of the NPF contribution to \(N_{\text{CCN}}\) and \(N_d\) based on a case study. Our study highlights the importance of fully consideration of both the environmental meteorological conditions and multiple sources (i.e. secondary and primary) to evaluate the NPF effect on clouds and the associated climate effects in polluted regions.”

Line 119-120: It is not clear if the SMPS measurements are done at ground level or the 260 m level. 

Re: The SMPS was deployed at ground level on about 8 m height from a 325 m meteorological tower. We have modified the sentence in lines 118-119, or as follows,

“The instruments during the campaign were deployed in a container at ground level (~8m on a meteorological tower).”

Line 213: Is a significant fraction of the pre-existing particles also from NPF? 

Re: This is a good point. There may be some pre-existing particles which could be
tracked from NPF. However, it is difficult to quantitatively calculate how many the particles are from NPF in the current study. But we think that the magnitude of the NPF-tracked particles in pre-existing aerosols should be much smaller than that particles number during NPF event. Further investigation warrants to conduct to clarify this probably combining with more instrument techniques in future.

Line 259: Why does the percentage of CCN enhancement increase significantly with supersaturation, but the percentage of CDNC enhancement is almost independent of vertical velocity?
Re: This is because that, when calculating CDNC, the supersaturation varies with the variations of $N_{CN}$. Under high $N_{CN}$, the water vapor competition effect will lead to lower supersaturation, which is smaller than that the constant supersaturations for calculating $N_{CCN}$. Roughly, the $N_d$ at $V$ of 0.3-3 m/s corresponds to the $N_{CCN}$ at $S$ of 0.1%-0.5%, within which the percentages of $\Delta N_{CCN}$ and the contributions of the NPF to $N_{CCN}$ don’t change much either. Some discussions have been included in lines 306-314 in the revised version.
“…With the increase of the $S$, the percentages of NPF-initiated $N_{CCN}$ and the contributions of the NPF to $N_{CCN}$ increased more significantly than that 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 on 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 $N_{CN}$, the water vapor competition effect will lead to lower $S_{max}$, which is smaller than that the constant $S$ for calculating $N_{CCN}$. Roughly, the $N_d$ at $V$ of 0.3-3 m/s corresponds to the $N_{CCN}$ at $S$ of 0.1%-0.5%, within which the percentages of $\Delta N_{CCN}$ and the contributions of the NPF to $N_{CCN}$ don’t change much either. The effect of water vapor competition will be further examined in the following section.…”

Line 344-345: I think these two numbers are not “contribution of NPF to $N_{CCN}$”, which should not exceed 100%.
Re: Yes, if we say “contribution of NPF to $N_{CCN}$”, the number should not exceed 100%. But, here, it refers to the increment in the percentage of the NPF-initiated $N_{CCN}$. We just have corrected the statements in lines 385-388, or as follows,
“As a result, a larger increment of $N_{CCN}$ is derived on clean NPF day, showing 37-80% and 15-41% increases percentage of $N_{CCN}$ from NPF on clean and polluted days respectively (Fig. 7b). As for $N_d$, on clean days are 22% and 37%, and 34% and 26% on polluted days under updraft velocity of 0.3 and 2.1 m s$^{-1}$.”
Figure 7. Comparison of the increments of (a) total particle number concentration ($N_{CN}$), and (b) CCN number concentration ($N_{CCN}$) and cloud dropet number concentration ($N_d$) between the two different typical NPF events.

Line 347: Why is the enhancement of CDNC so similar in polluted NPF days and clean NPF days, while the enhancement of CCN is quite different?  
Re: According to the Fig.7a, as it is more conductive for the formation of NPF and more Aitken mode particles increased on clean day, the increment percentage of $N_{CCN}$ on clean day is much larger than that on polluted day. While the $N_{CN}$ in polluted atmosphere is high, the impact of water vapor competition on $N_d$ is more significant on polluted day. The relevant statements are in lines 389-393, or see as follows, “...The result just further illustrates that the effect of water vapor competition on $N_d$ under high $N_{CN}$ in 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...”

This manuscript needs to be carefully edited by a native speaker to improve the English writing. 
Re: the manuscript has been corrected and edited carefully.