RESPONSES TO REVIEWERS' COMMENTS

Dear Editor,

We are very pleased to submit the revised manuscript entitled "Evaluation of the contribution of new particle formation to cloud droplet number concentration in urban atmosphere" for possible publication in journal of ACP.

I'd like to thank you for your attention for this matter. I also would like to thank the reviewers for their insightful and constructive comments and suggestions. The paper has been revised according to the reviewers' recommendations. Below please find our detailed point-by-point responses (in blue) to the reviewers' comments (in black) to the manuscript. We believe that, after a major revision by considering the major concerns from the reviewer carefully, the paper has been improved greatly.

Yours sincerely,

Fang Zhang

On behalf of all authors

Review#2

The manuscript presents observations on the effects of new particle formation (NPF) on cloud condensation nuclei (CCN) concentrations in urban Beijing. These observations are used to quantify the limiting effects of extremely high CCN concentrations on cloud activation. The topic is scientifically relevant and suitable for ACP. However, there are some issues that need to be solved before the manuscript can be accepted for publication.

General comments

The paper focuses only on 7 "typical" NPF events, which is a small data set. Potential issues related to the small sample size should be discussed and limitations should be noted when drawing conclusions. Regarding the small sample size, it would be important to justify why these seven days were selected and what makes them typical. There are also case studies focusing on "typical" single days, which is generally fine, but again these should be justified.

Re: Thanks a lot for pointing this. Regarding to the small size of the dataset, we just have included some statements in the revised version to clarify this limitation (lines 496-500) or as follows. When discussing the results, we also used some attributive words to emphasize that the results only represent the case during the observation periods in urban Beijing. Regarding to the reviewers concerns on the selection of the "typical" NPF events, we also included some new statements to justify the issues, please see the lines 264-271 or as follows,

"... According to Dal Maso 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 time scale (e.g. local sources from vehicle or cooking emissions), but they do not show the "banana" shape. Therefore, those cases with typical "banana" shape (7 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...."

"... 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, warrant to verify and refine our results, so as to parameterize the impact of NPF events on cloud, precipitation, and radiative forcing in models....."

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, Boreal Environ. Res., 10, 323–336, 2005.

Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of

hygroscopicity measurements, Atmos. Chem. Phys., 15, 13071–13083, doi:10.5194/acp-15-13071-2015, 2015.

Contrasting cloud droplet number concentrations based on a constant maximum supersaturation and updraft velocity seems like comparing apples and oranges. It is well known CCN compete for water vapor during cloud activation and for adiabatically rising air parcels this is one of the main factors determining the maximum supersaturation. Why would using a prescribed updraft velocity be better than prescribed maximum supersaturation in calculating CDNC? Surely, both updraft velocity and cloud supersaturation are parameters that are difficult to measure in the actual environment. While updraft velocity has its use in the modeling community, CDNC at a fixed maximum supersaturation is a useful parameter to compare with observations.

Re: Usually, the measurement of CCN number concentration was under a preset fixed supersaturation in the CCN counter instrument. When evaluating the cloud droplet number concentration (N_d) at the prescribed updraft velocity, the maximum supersaturation (S_{max}) in the environment at each moment is always changing due to the water vapor competition effect. So, different from the CCN number concentration, the number concentration of cloud droplets not only depend on the particle number size distribution and chemical composition at each moment but also on the updraft velocities. Therefore, using the prescribed updraft velocity can reflect the number of cloud drops in adiabatic ascending clouds more realistically than the constant supersaturation. Some statements have been included in the revised text to clarify this, see lines 66-74, or as follows,

"... 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)....."

Nenes, A. and Seinfeld, J. H.: Parameterization of cloud dropletformation in global climate models, J. Geophys. Res, 108, 4415, https://doi.org/10.1029/2002JD002911, 2003.

Hudson, J. G. , Noble, S., and Tabor, S.: Cloud supersaturations from CCN spectra hoppel minima. Journal of Geophysical Research Atmospheres, 120(8), 3436–3452. https://doi.org/10.1002/2014JD022669, 2015.

Some calculations and choices require better explanations. Specific comments related to this and other issues are given below.

Specific comments

Lines 48-49: contributions of NPF on aerosol (number concentration) needs a better explanation. Are the numbers from the reference (Merikanto et al., 2010)? Is this for boundary layer? Are there any other references (e.g., urban or China)?

Re: we have included more explanations and reference of studies in urban area of China in the revised main text in lines 56-65, or see as follows,

"... The NPF events was 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 the NPF contributed about 76% of the total fine particle number concentrations 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., 2011). In reality, the field studies have shown that these fine particles produced from NPF can subsequently turn into an enhancement in N_{CCN} at cloud-relevant supersaturations (Kalkavouras et al., 2017; Peng et al., 2014; Wu et al., 2015; Ma et al., 2016; 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)....."

Shi, J., Khan, A., Harrison, R.: Measurements of ultrafine particle concentration and size distribution in the urban atmosphere, Science of the Total Environment, 235(1-3):51-64,doi:10.1016/S0048-9697 (99)00189-8, 1999.

Stanier, C., Khlystov, A., Pandis, S.: Ambient aerosol size distributions and number concentrations measured during the Pittsburgh Air Quality Study (PAQS), Atmospheric Environment, 38(20):3275-3284, doi:10.1016/ j.atmosenv.2004.03.020, 2004.

Kulmala, M. and Kerminen, V. M.: On the formation and growth of atmospheric nanoparticles, Atmos. Res., 90, 132–150, doi:10.1016/j.atmosres.2008.01.005, 2008.

Wu, Z. J., Hu, M., Yue, D. L.: Evolution of particle number size distribution in an urban atmosphere during episodes of heavy pollution and new particle formation. Sci China Earth Sci, 2011, 54, doi:10.1007/s11430-011-4227-9,2011.

Fan, J., Leung, R., Rosenfeld, D., Chen, Q., Li, Z., Zhang, J., and Yan, H.: Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds, P. Natl. Aacad. Sci. USA, 110, E4581–E4590, https://doi.org/10.1073/pnas.1316830110, 2013.

Li, Z., Niu, F., Fan, J. et al. Long-term impacts of aerosols on the vertical development of clouds and precipitation. Nature Geosci 4, 888–894 (2011). https://doi.org/10.1038/ngeo1313,doi:10.1038/ngeo1313, 2011.

Kalkavouras, P., Bossioli, E., Bezantakos, S., Bougiatioti, A., Kalivitis, N., Stavroulas, I., Kouvarakis, G., Protonotariou, A. P., Dandou, A., Biskos, G., Mihalopoulos, N., Nenes, A., and Tombrou, M.: New particle formation in the southern Aegean Sea during the Etesians: importance for CCN production and cloud droplet number, Atmos. Chem. Phys., 17, 175–192, https://doi.org/10.5194/acp-17-175-2017, 2017.

Peng, J. F., Hu, M., Wang, Z. B., Huang, X. F., Kumar, P., Wu, Z. J., Guo, S., Yue, D. L., Shang, D. J., Zheng, Z., and He, L. Y.: Submicron aerosols at thirteen diversified sites in China: size distribution, new particle formation and corresponding contribution to cloud condensation nuclei production, Atmos. Chem. Phys., 14, 10249–10265, doi:10.5194/acp-14-10249-2014, 2014.

Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements, Atmos. Chem. Phys., 15, 13071–13083, doi:10.5194/acp-15-13071-2015, 2015.

Ma, N., Zhao, C., Tao, J., Wu, Z., Kecorius, S., Wang, Z., Größ, J., Liu, H., Bian, Y., Kuang, Y., Teich, M., Spindler,

G., Müller, K., van Pinxteren, D., Herrmann, H., Hu, M., and Wiedensohler, A.: Variation of CCN activity during new particle formation events in the North China Plain, Atmos. Chem. Phys., 16, 8593–8607, doi:10.5194/acp-16-8593-2016, 2016.

Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu, B.: Aerosols and boundary-layer interactions and impact on air quality. Natl. Sci. Rev., 4, 810–833, doi:10.1093/nsr/nwx117, 2017.

Zhang, F., Ren, J., Fan, T., Chen, L., Xu, W., Sun, Y.: Significantly enhanced aerosol CCN activity and number concentrations by nucleation‐initiated haze events: A case study in urban Beijing. J. Geophys. Res.-Atmos., 124. doi:10.1029/ 2019JD031457, 2019

Wiedensohler, A., Chen, Y. F., Nowak, A., Wehner, B., Achtert, P., Berghof, M., Birmili, W., Wu, Z. J., Hu, M., Zhu, T., Takegawa, N., Kita, K., Kondo, Y., Lou, S. R., Hofzumahaus, A., Holland, F., Wahner, A., Gunthe, S. S., Rose, D., Su, H., and Pöschl, U.: Rapid aerosol particle growth and increase of cloud condensation nucleus activity by secondary aerosol formation and condensation: A case study for regional air pollution in northeastern China, J. Geophys. Res., 114, D00G08, doi:10.1029/2008JD010884, 2008.

Lines 112-116 and 187-191: were there specific conditions for identifying/classifying NPF events? Specifically, how the seven NPF cases (Fig. 1; line 190) were selected? The authors write that "While non-NPF events may also have sudden increases of nucleated particles at a short time scale, but they do not show further growth" (lines 115-116), so does this mean that the focus is on particle growth rather than NPF?

Re: According to Dal Maso 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 time scale (e.g. local sources from vehicle or cooking emissions), but they do not show the "banana" shape. Therefore, those cases with typical "banana" shape (7 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. This has also been revised and clarified in the revised text, see lines261-267, or as follows,

"... According to Dal Maso 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 time scale (e.g. local sources from vehicle or cooking emissions), but they do not show the "banana" shape. Therefore, those cases with typical "banana" shape (7 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...."

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, Boreal Environ. Res., 10, 323–336, 2005.

Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of

Section 2.2: this section could be clarified. Mixing the growth of particles (dry size) and droplets (wet size) is confusing. This includes also the critical droplet/dry size. There is also minimum particle size, which is somehow mixed with critical diameter (Eqs 1 and 2). Eqs 1 and 2 have also unexplainable variables. Which species are "organic and inorganic" (line 145) and what are their hygroscopic parameters?

Re: In the revised version, we use "D_p" to represent the dry particle diameter and "D_c" to represent the critical dry particle diameter. The organic species and inorganic species and their hygroscopic parameters are added and summarized in Table 1. Therefore, this paragraph in section 2.2 have been revised as follows, or see lines 135-165:

"…According to the hygroscopic growth process of particles described by Köhler theory (Köhler et al., 1936), the particles with the 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 κ-Köhler theory (Petters and Kreidenweis, 2007), which simply describe the approximate relationship between the D_c with the critical supersaturation (S_c), is applied as follows, when $\kappa > 0.1$:

$$\kappa = \frac{4A^3}{27D_c^3 ln^2 S_c},\ A = \frac{4\sigma_w M_w}{RT\rho_w} \quad (1)$$

where M_w is the molecular weight of water (M_w = 0.018015 kg mol^{-1}), ρ_w is the density of water ($\rho_{w=}$ = 997.1 kg m^{-3}), T is the parcel temperature (T = 298.15 K), where σ_w is the droplet surface tension at the point of activation (σ_w = 0.072 J m^{-2}) and R is the universal gas constant (R = 8.315 J K^{-1} mol^{-1}). κ 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 κ 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 volume fraction of organic and inorganic, according to the following equation:

$$\kappa_{chem} = \sum_i \varepsilon_i \kappa_i \quad (2)$$

where κ_i and ε_i are the hygroscopic parameter and volume fraction for each individual (dry) component in the mixture, respectively. The κ value and density (ρ) of each species used in the calculation are given in Table 1, which is referred from Petters and Kreidenweis (2007) and Topping (2005).

Table 1. Densities of different chemical species and their κ measured by the laboratory

Species	NH_4NO_3	$(NH_4)_2SO_4$	NH_4HSO_4	H_2SO_4	POA*	SOA*	BC
ρ (kg m-3)	1720	1769	1780	1830	1000	1400	1700
κ	0.58	0.48	0.56	1.19	0	0.09	0

POA* refers to primary organic aerosol and SOA* refers to secondary organic aerosol

In the equation (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:

$$CCN(D_c) = \int_{D_c}^{550} n(logD_p)dlogD_p \quad (3)$$

where $n(logD_p)$ is the particle number that correspond to each particle size bin $dlogD_p$ in the aerosol number size distribution.…"

Köhler, H.: The nucleus in and the growth of hygroscopic droplets, Transactions of the Faraday Society, 32: 1152-1161, doi:10.1039/tf9363201152, 1936.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, doi:10.5194/acp-7-1961-2007, 2007.

Section 2.3: this section could be clarified. dw/dt in Eq. 4 needs to be explained. In addition, it is not clear how maximum supersaturation is solved from this equation. Supersaturation has different symbols (s and S), and Eq. 5 has unexplainable variables and index i. It would be useful to give more details about "empirical values of cloud updraft velocity" (line 170) to justify the selected updraft velocity range.

Re: Thanks a lot for the careful check and review on this work. We have just revised and corrected the Section very carefully, including adding more words to explain the variavles in the equations, unifying the symbols of "S", etc. Several references have also be included to clarify the empirical values of cloud updraft velocity. Please see the corrections as follows,

Lines 175-181:

"...while the $\frac{dw}{dt}$ denotes the water condensation rate during the aerosol activation and subsequent growth processes. which is shown in detail in Eqs (5). And the $\frac{ds}{dt}$ express the growth rate of supersaturation, when it is equal to 0, the supersaturation reaches the maximum value.

$$\frac{dw}{dt} = \frac{\pi}{2}\rho_w \int_0^S D_p^2 \frac{dD_p}{dt} n^S(S')dS' \quad (5)$$

where ρ_w is the density of water. $n^s(S')dS'$ is the number concentration of particles activated between S' and $S' + dS$...."

Lines 182-187:

"...Nenes et al.(2002) used a sectional representation of the CCN spectrum (i.e. particle number supersaturation distribution $n^s(s')$) and total number of particles with S_c smaller than S, $F^S(S)$, which is given by

$$F^S(S_x) = \int_0^{S_x} n^S(S')dS' \quad (6)$$

Where the S_x 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 $F^S(S_x)$ can be calculated by the integration of $n^S(S')$ from the lower limit 0 to upper limit S_x....."

Lines 191-199:

"…In this study, we used the PNSD, chemical components, and empirical values of cloud updraft velocity to determine the S_{max} and N_d during NPF days in urban Beijing. Owing to that the direct measurement of cloud-scale updraft velocity in the atmosphere is almost impossible, the prescribed updraft velocity used in this study is referred from previous studies. Generally, the updraft velocities are reported very small (Martin et al., 1994) and range from 0.1 to 1.0 m/s in stratocumulus and cumulus clouds in remote or marine boundary layer (Meskhidze et al., 2005; Morales et al., 2010). The vertical updraft velocities were derived varying from 0.3 to 3 m/s (Zheng et al., 2015), which are typical for cumulus and convective clouds in summer of north China and thus was selected and applied in this study…."

Nenes, A., Chan, S., Abdul-Razzak, H., Chuang, P., and Seinfeld, J. H.: Kinetic limitations on cloud droplet formation and impact on cloud albedo, Tellus 53B, 133–149, doi:10.3402/tellusb.v53i2.16569, 2001.

Nenes, A. and Seinfeld, J. H.: Parameterization of cloud dropletformation in global climate models, J. Geophys. Res, 108, 4415, https://doi.org/10.1029/2002JD002911, 2003.

Martin, G., Johnson, D.: The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds.[J]. Journal of the Atmospheric Sciences, doi:10.1175/1520-0469(1994)051<1823:TMAPOE>2.0.CO;2,1994.

Morales, R. and Nenes, A.: Characteristic updrafts for computing distribution-averaged cloud droplet number and stratocumulus cloud properties, J. Geophys. Res., 115, D18220, https://doi.org/10.1029/2009JD013233, 2010.

Zheng, Y. T.,Rosenfeld, D.,Li, Z. Q.: Satellite Inference of Thermals and Cloud-Base Updraft Speeds Based on Retrieved Surface and Cloud-Base Temperatures[J], Journal of the Atmospheric Sciences,2015,72(6), doi: 10.1175/JAS-D-14-0283.1,2015.

Figure 1: it looks like the smallest particles (below 20 nm) are missing; is this a measurement artefact or a real observation? Does this cause difficulties in identifying NPF events? At least nucleation mode (10-25 nm) particle concentration would be underestimated.

Re: In this study, the measured particle size range by SMPS is 11.3 -552.3 nm. Although the ultrafine particles with size smaller than 10 nm are missing, the NPF events can be observed and identified clearly. Also, the CCN-size particles are much larger (usually >50 nm) at cloud-relevant supersaturations in the studied region. Therefore, the lack of measurement of small particles below 10 nm will not impact our results.

On line 195 it is stated that "The variation of N_{CCN} seems to be more consistent with that of N_{CN} than N_d", but is this true? Please clarify the text and also show some

evidence (for example correlation coefficients) to support this.

Re: It can be verified by comparing the correlations between N_{CN} and N_{CCN} at supersaturations of 0.6% and 0.8% and the correlations between N_{CN} and N_d under updraft vertical velocities of 2.1 m s^{-1} and 3.0 m s^{-1}. We can see the Fig. 4 of the text, the N_{CCN} and N_{CN} were obviously linear correlated, but the correlation between N_d and N_{CN} was non-linear. The correlation coefficients are also in Fig. 4. The relevant statements are in lines 268-273, 316-322 or as follows,

"…It exhibits that the NPF event drives the variation of N_{CCN} and N_d, showing that the occurrence of NPF events as an important source of CCN. The variation trend of N_{CN} is more correlated with that of N_{CCN} than N_d (also see Fig. 4, Table S5). This is because that the N_{CCN} was calculated based on a constant S rather than refer to the availability of water vapor, while the calculation of N_d is based on the S_{max} that can reach in the real atmosphere at a given updraft velocity…"

"…Fig. 4 shows the scatter plots of correlations between N_{CN} and N_{CCN} at supersaturations of 0.6% and 0.8% and the correlations between N_{CN} and N_d under updraft vertical velocities of 2.1 m s^{-1} and 3.0 m s^{-1}. The N_{CCN} and N_{CN} were obviously linearly related, but the correlation between N_d and N_{CN} was non-linear. When shown as the average values with error bars, the N_d increase linearly as N_{CN} increase when the N_{CN} is below 15000, then the N_d began to decrease with the further increase of N_{CN}. This has been presented in previous studies (Nenes et al., 2001; Ramanathan et al., 2001; Sullivan et al., 2016), and was believed to be caused by the water vapor competition of the aerosol particles…."

Nenes, A., Chan, S., Abdul-Razzak, H., Chuang, P., and Seinfeld, J. H.: Kinetic limitations on cloud droplet formation and impact on cloud albedo, Tellus 53B, 133–149, https://doi.org/10.3402/tellusb.v53i2.16569, 2001.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle, Science, 294, 2119–2124, https://doi.org/10.1126/science.1064034, 2001.

Sullivan, S. C., Lee, D., Oreopoulos, L., and Nenes, A.: The role of updraft velocity in temporal variability of cloud hydrometeor number, P. Natl. Acad. Sci. USA, 113, 5781–5790, https://doi.org/10.1073/pnas.1514043113, 2016.

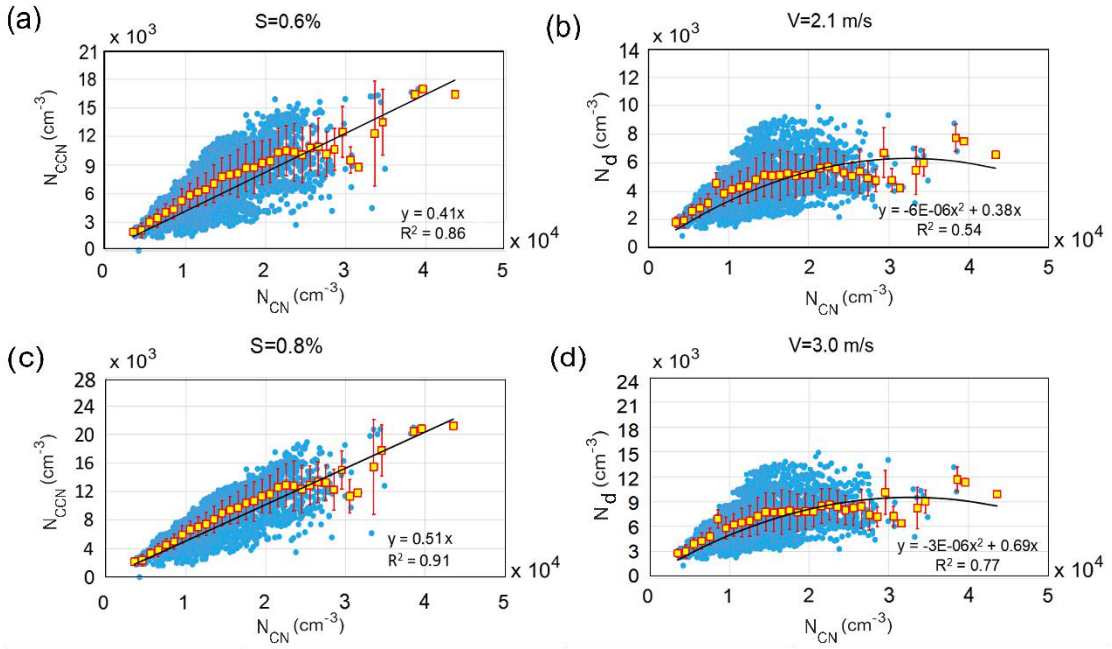

Figure 4. Scatter plots of correlation between total number concentration (N_{CN}) and CCN number concentration (N_{CCN}) at supersaturation of (a) 0.6% and (c) 0.8% respectively. Scatter plot of correlation between N_{CN} and cloud droplet number concentration (N_d) at updraft vertical velocity of (b)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

Table S5. The correlation between N_{CN} and N_{CCN}, N_{CN} and N_d

Updraft velocities V	R of N_{CN} and N_d (at V)	The S_{max} corresponding to V	R of N_{CN} and N_{CCN} (for S_{max})
0.3 m/s	0.51	0.23%	0.61
0.9 m/s	0.69	0.37%	0.75
1.5 m/s	0.75	0.47%	0.79
2.1 m/s	0.77	0.59%	0.88
3.0 m/s	0.81	0.73%	0.93

R is the correlation coefficients

In general, it would be better to write that N_{CCN} is based on a constant maximum supersaturation rather than refer to the availability of water vapor (lines 196-197).

Re: We have revised in the main text in lines 271-274, , or as follows,

"... This is because that the N_{CCN} was calculated based on a constant S rather than refer to the availability of water vapor, while the calculation of N_d is based on the S_{max} that can reach 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} ...."

Calculation of the effect of NPF on CCN and clouds should be clarified. Please explain "decoupling time" (line 202): how is it defined and what does it mean in

practice. Is there a specific definition for t_{start} (line 203) and how is it related to the start of the period from 12:00 to 16:00 to estimate the change of CCN during NPF events (line 204)? Also, using the constant time period (12:00-16:00) should be justified; this works only if the events start at the same time. What are the averaging time periods for calculating CCN concentrations before and during NPF events (line 207)?

Re: The "decoupling time" is a time when CCN number concentration begin to increase due to the newly formed nucleation particles grow into CCN relevant size, and it was defined by Kalkavouras et al (2019). We used the method and definition of Kalkavouras et al (2019) to determine the time when NPF begins to impact both N_{CCN} and N_d. And we named the time as "$t_{start,CCN}$" instead of "t_{dec}" in the revised version. In addition, the time when NPF stop impacting the N_{CCN} is named as $t_{end,CCN}$. t_{start} is the time when NPF event occurrs, which is usually several hours earlier than $t_{start,CCN}$ due to that it takes time for newly formed nucleation particles to grow to the relevant size of CCN. Since the $t_{start,CCN}$ are different for different NPF cases (Table S1, S2), in the revision, the constant time period (12:00-16:00) was no longer used to evaluate the enhancements of CCN during NPF events. The details of the method are given in Section 2.4, or see lines 200-235, or as follows:

"…**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 (ΔN_{CCN} or ΔN_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_{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_{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 $t_{start,CCN}$ and $t_{end,CCN}$ respectively, the time series of N_{CCN} was firstly divided by the N_{CCN} at t_{start} at each prescribed supersaturation, to derive the normalized time series of N_{CCN}, denoted as R_s. The equation is written as follows,

$$R_S = \frac{CCN_S}{CCN_{S,t_{start}}} \quad (8)$$

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)…."

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Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements, Atmos. Chem. Phys., 15, 13071–13083, doi:10.5194/acp-15-13071-2015, 2015.

Ma, N., Zhao, C., Tao, J., Wu, Z., Kecorius, S., Wang, Z., Größ, J., Liu, H., Bian, Y., Kuang, Y., Teich, M., Spindler, G., Müller, K., van Pinxteren, D., Herrmann, H., Hu, M., and Wiedensohler, A.: Variation of CCN activity during new particle formation events in the North China Plain, Atmos. Chem. Phys., 16, 8593–8607, doi:10.5194/acp-16-8593-2016, 2016.

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Zhang, F., Ren, J., Fan, T., Chen, L., Xu, W., Sun, Y.: Significantly enhanced aerosol CCN activity and number concentrations by nucleation‐initiated haze events: A case study in urban Beijing. J. Geophys. Res.-Atmos., 124. doi:10.1029/ 2019JD031457, 2019

Fan, X., Liu, J., Zhang, F., Chen, L., Collins, D., Xu, W., Jin, X., Ren, J., Wang, Y., Wu, H., Li, S., Sun, Y., and Li, Z.: Contrasting size-resolved hygroscopicity of fine particles derived by HTDMA and HR-ToF-AMS measurements between summer and winter in Beijing: the impacts of aerosol aging and local emissions, Atmos. Chem. Phys., 20, 915–929, https://doi.org/10.5194/acp-20-915-2020, 2020.

Table S1. The critical time in evaluating CCN enhancements on seven NPF days

NPF case	t_{start}	t_{CCN}	$t_{end,CCN}$
25 May	09:00	12:40	03:00^{+1}
27 May	11:00	14:00	17:10
01 June	09:00	11:20	20:10
02 June	12:00	13:30	16:20
07 June	09:00	12:00	17:10
10 June	09:00	14:10	23:30
11 June	11:00	11:50	18:00

Table S2. The critical time in evaluating N_d enhancements on seven NPF days

NPF case	t_{start}	t_{Nd}	$t_{end,Nd}$
25 May	09:00	13:40	04:00^{+1}
27 May	11:00	14:00	00:00
01 June	09:00	11:10	12:00^{+1}
02 June	12:00	01:30^{+1}	12:00^{+1}
07 June	09:00	11:25	12:00^{+1}
10 June	09:00	12:05	18:35
11 June	11:00	16:30	21:00

$^{+1}$ means the next day

Calculation of the effect of NPF on CCN and clouds does not account for the changes in background conditions (except the evening traffic emissions at 16:00). This should be examined and explained in the manuscript. For example, would there be a change in CCN concentration during non-event days?

Re: as the reviewer commented, the changes in background conditions (like primary emission and the evolution of the PBL) will impact on aerosol particles, and consequently on its CCN number concentrations. But as shown in Fig. 2, the time series of N_{CN} show that NPF is the main factor impacting on the variation in N_{CN}. And the changes in N_{CN} before and after the NPF event is relatively small during the period of non-NPF events. Some revisions and statements have been included in the text (lines 241-252), or as follows,

"…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; Altstädter, 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…."

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Altstädter, B., Platis, A., Wehner, B., Scholtz, A., Wildmann, N., Hermann, M., Käthner, R., Baars, H., Bange, J., and Lampert, A.: ALADINA – an unmanned research aircraft for observing vertical and horizontal distributions of ultrafine particles within the atmospheric boundary layer. Atmos. Meas. Tech., 8, 1627–1639, doi:10.5194/amt-8-1627-2015, 2015.

Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu, B.: Aerosols and boundary-layer interactions and impact on air quality. Natl. Sci. Rev., 4, 810–833, doi:10.1093/nsr/nwx117, 2017.

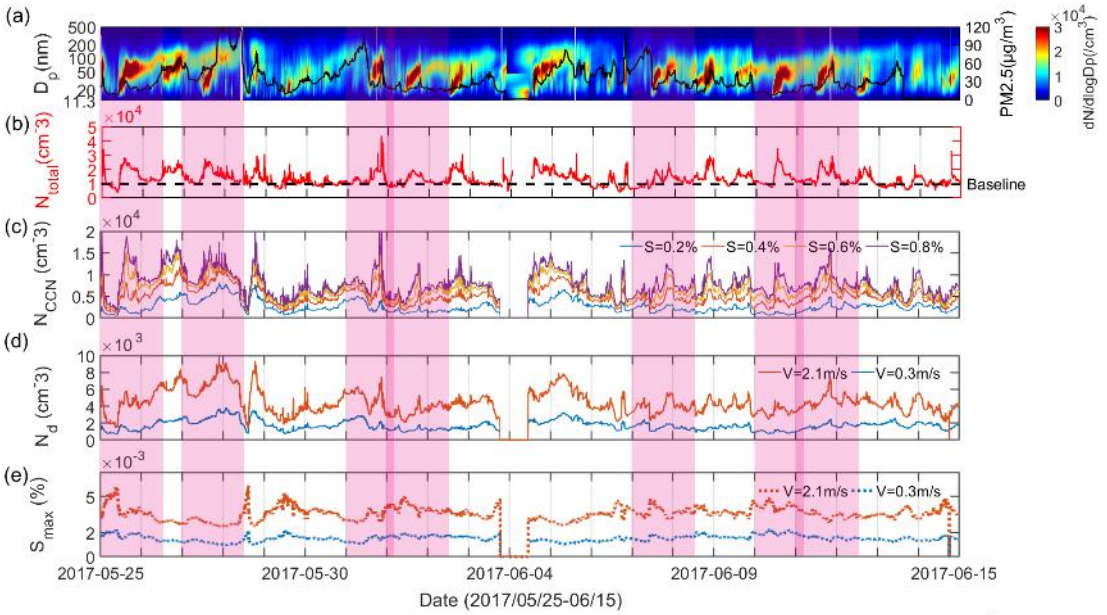

Figure 2. Time series of (a) particle number size distribution (PNSD) (the selected 7 typical NPF events are marked in pink shadow), (b) the total particle number concentration (N_{total}), (c) CCN number concentration (N_{CCN}), (d) cloud droplets number concentration (N_d) and (e) the maximum supersaturation (S_{max}) from 25 May to 15 June 2017.

Where is it shown that NPF drives the variation of N_{CCN} (line 210)? What is the role of background particles and primary emissions (Sect. 3.5) for variation of CCN concentration?

Re: As we stated above, the NPF is the main factor impacting on the variations of N_{CN}., and thus N_{CCN} and N_d. During the studied period, we observed that the variations of the concentrations of the background particles during non-NPF or clean periods were insignificant (Fig. 2). However, the particles from primary emissions also contribute to N_{CCN} and N_d. Our investigation shows that about 70% N_{CCN} and N_d are

from background pre-existing particles including 10%-20% N_{CCN} and N_d are from primarily emitted particles. Some statements have been included in the text to clarify this issues (see lines 283-285, 306-306 and 450-452), or see as follows:

"And the rest (about 63-76%) of CCN are from the other sources or pre-existing particles, which is much larger than that derived in remote Finokalia, Crete, Greece by Kalkavouras et al (2019)…We also show that the NPF contributes about 30% to the total N_d during the studied period in urban Beijing. And the rest (about 70%) of cloud droplet are from the other sources or pre-existing particles…."

"…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… For N_{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…"

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Calculation of the effect of NPF on cloud droplet number should be clarified. The explanation in lines 237-246 is confusing. Adiabatic rise does not produce water vapor (line 239) but has an effect on vapor pressure (or saturation). I don't think that cloud droplets "feel" particles (line 244), but they will compete with other cloud droplets about the available water vapor. Terminology could be similar with the section focusing on the effect of NPF on CCN. Is t_{start} the same in all cases (lines 203, 247 and 333)? Is t_{Nd} always 12:00 (line 246)? Is the end time case-dependent, why? If the event end time are different, this will have an effect on CCN and cloud droplet enhancements.

Re: As the reviewer commented, 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 believe the method has been clearly addressed after the major revision. The details of the method are given in Section 2.4, or see lines 201-235, or as follows:

"…**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 (ΔN_{CCN} or ΔN_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_{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_{start}.

Since there need 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 $t_{start,CCN}$ and $t_{end,CCN}$ respectively, the time series of N_{CCN} was firstly divided by the N_{CCN} at t_{start} at each prescribed supersaturation, to derive the normalized time series of N_{CCN}, denoted as R_s. The equation is written as follows,

$$R_S = \frac{CCN_S}{CCN_{S,t_{start}}} \quad (8)$$

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)…."

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Fan, X., Liu, J., Zhang, F., Chen, L., Collins, D., Xu, W., Jin, X., Ren, J., Wang, Y., Wu, H., Li, S., Sun, Y., and Li, Z.: Contrasting size-resolved hygroscopicity of fine particles derived by HTDMA and HR-ToF-AMS measurements between summer and winter in Beijing: the impacts of aerosol aging and local emissions, Atmos. Chem. Phys., 20, 915–929, https://doi.org/10.5194/acp-20-915-2020, 2020.

Variance analysis (line 261-265) needs a brief explanation in the main text (now there is nothing in the main text and the equations in the supplementary material seem to be incorrect). I suggest either presenting the method in the main text or removing this part from the manuscript.

Re: Thanks a lot for the reviewer's suggestion. After a careful consideration, we just removed this analysis in the main text since it is not closely relevant to the research topic.

Section 3.3 focuses on the effect of water vapor competition (or depletion), which is a known phenomenon related to cloud activation. The first paragraph (lines 271-287) could be reformulated for clarity (technical corrections listed below). Terms “with and without the water vapor depletion” (line 294) are misleading, because these droplet numbers are based on fixed updraft velocity and constant supersaturation, respectively. The depletion effect is included also in the latter approach because its constant supersaturation is taken from calculations “with water vapor depletion”. Another reason for not to compare these two approaches is that the constant updraft velocity method accounts also for hygroscopicity variations. For these reasons I would not use term water vapor depletion/competition in this context.

Re: this paragraph has been clarified and revised. The terms like “with and without the water vapor depletion”, and “water vapor depletion” which the reviewer mentioned have been removed. In reality, in the calculation of N_{CCN} and N_d, the changes of hygroscopic parameters were taken into account, as the κ-Köhler theory were used in both of them. The revised descriptions and corrections can be found in the revised version in lines 323-325, 330-332 or as follows,

“... 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 to a certain number. This is fully suggested by the difference between the calculated N_{CCN} using the constant S and the N_d using the variable S_{max} in the air parcels. Because

in the actual environment, it is often unable to achieve the sufficient supersaturation compared to the prescribed ones that are preset in the instrument...."

"... Therefore, although NPF events may strongly increase N_{CCN}, the formed N_d are eventually limited by water vapor competition which determines the S_{max} that varies in the cloud. The S_{max} is related to the cloud formation dynamics and the aerosol levels in the region...."

Section 3.5 focuses on primary emissions, but a few things need to be clarified. First, please explain how these were identified as primary particles? Secondary (SOA) and primary (POA) organic aerosols are shown in Fig. 9, and there are high concentrations of SOA. How were SOA and POA calculated? This information is currently completely missing and must be added. Isn't there any background aerosol between 18:00 and 21:30 (lines 370-371) or at any other time? How was the growth rate of the newly formed particles calculated (line 385)? The method for separating PNSD of primary emissions (lines 386-390) needs a better explanation. It seems to be based on the fitted modes, so please explain why the three modes were selected (there could be more than three) and justify their origin (no background aerosol and no effect from boundary layer dynamics?). Why weren't the modes tracked from the beginning of NPF event? This could have shown their origin. Better justification is needed to convince readers that Figs 9e and 9f show PNSDs of NPF and primary emissions, respectively.

Re: Regarding to the identification of the POA and SOA, we have included more words to describe the source appointment method of PMF for separating the primary and secondary aerosols in the revision,see lines 411-418, or as follows,

".. 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 a 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 OA 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 support information of Liu et al., (2021).

…"

We also have added more words to explain how we select three modes to fit the PNSD of NPF-tracked and primary aerosols in the revised text, please see lines 400-420, or as follows,

"…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 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±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 ~50-60 nm during the rush hour period. While, the primary particles from vehicles or cooking are generally with a smaller size (~30 nm) than the NPF-tracked particles mode and accumulation mode (~100-120 nm) according to 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 ~50 nm that is related to the NPF event, and two minor peaks in Aitken (~30 nm) and accumulation (~100-120 nm) mode (Fig. 8e, f) that are associated with the primary vehicle or cooking emissions. Fig. 8g and Fig. 8h show the separated PNSD of the NPF-related and primary aerosols respectively. Then the increment of N_{CCN} and N_d from NPF are obtained from the PNSD of NPF mode, and the increment of N_{CCN} and N_d from primary emissions are obtained by subtracting the increment of N_{CCN} and N_d by NPF from the total increment of N_d…."

About the potential impact from the background particles and variations of boundary layer, some more statements and discuss have been included in the revised text, see lines 241-252, and lines 461-465, or see as follows:

" 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; Altstädter, 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."

"…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)…."

The calculation of the growth rate of the newly formed particles have been added in lines 422-425, or as follows,

"The size mode of the newly formed particles during the rush hour is estimated

by applying a growth rate of 3.2±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 ~50-60 nm during the rush hour period."

Kerminen, V. M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi, E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala, M., and Petäjä, T.: Cloud condensation nuclei production associated with atmospheric nucleation: a synthesis based on existing literature and new results, Atmos. Chem. Phys., 12, 12037-12059, doi:10.5194/acp-12-12037-2012, 2012.

Brines, M., Dall'Osto, M., Beddows, D. C. S., Harrison, R. M., Gómez-Moreno, F., Núñez, L., Artíñano, B., Costabile, F., Gobbi, G. P., Salimi, F., Morawska, L., Sioutas, C., and Querol, X.: Traffic and nucleation events as main sources of ultrafine particles in high-insolation developed world cities, Atmos. Chem. Phys., 15, 5929–5945, https://doi.org/10.5194/acp-15-5929-2015, 2015.

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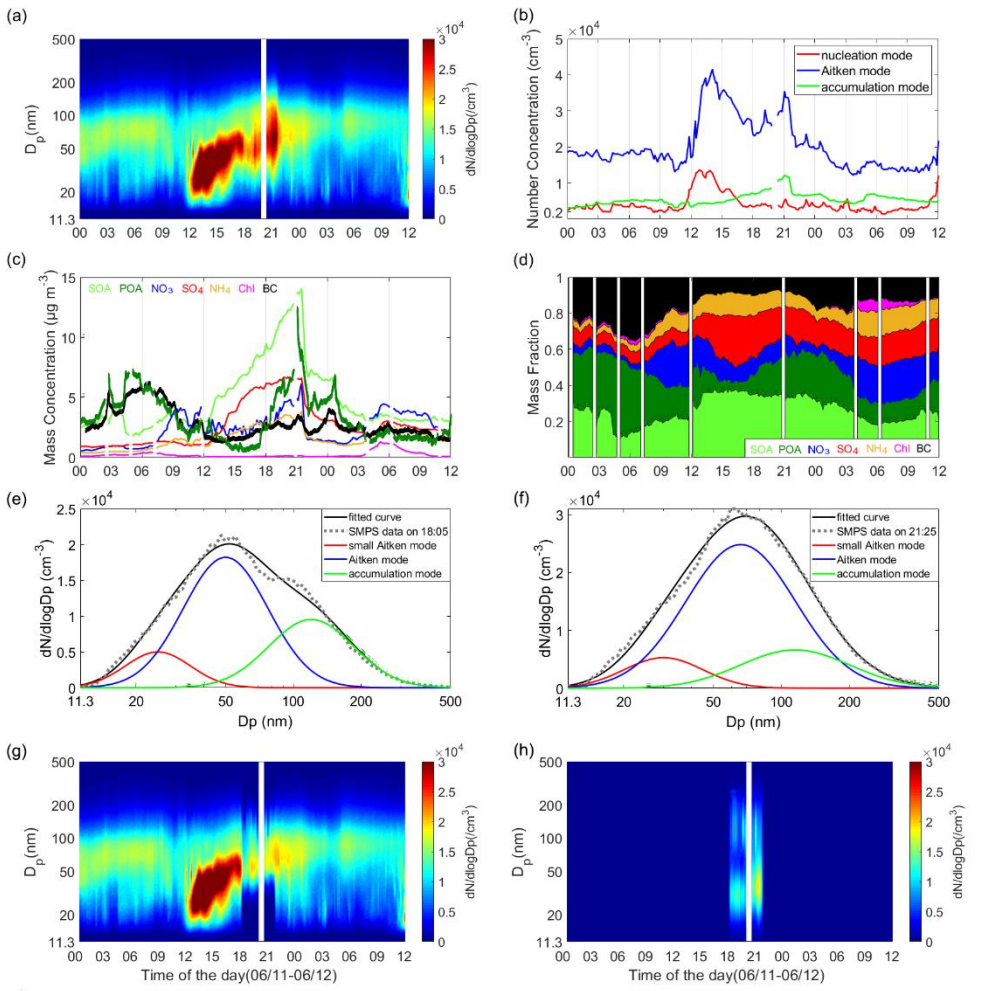

Figure 8. Diurnal variations of the (a) aerosol size distribution, (b) particle number concentrations for different size modes, (c) mass concentrations of aerosol chemical

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.

Technical corrections

Below are line numbers and the part of the text that could be improved/fixed/clarified is quoted. Some of these appeared in the text more than once, but only the first line is listed here.
Re: Thanks a lot for the careful check and review on this work. we have carefully checked your comments and modified the relevant expression in the main text or see in lines in the revised version respectively.

1: "cloud droplet" or maybe cloud droplet [number] concentration?
Re: It refers to cloud droplet number and we have revised it with "cloud droplet number concentration" in line 1 in the revised version.

19: "markedly reduction"?
Re: The "markedly" is a adverb and we have replaced it with "significant reduction" in line 29 and 471 in the revised version.

28: "particles are very low"
Re: we have revised this sentence into "when there are few pre-existing background aerosol particles" in lines 38 in the revised version.

39: "a lot of researchers"
Re: "a lot of" is removed in the sentence (lines 49-50).

39: "parametric model"
Re: we have replaced the term with "numerical activation models" in line 50 in the revised version, and it also appears in line 152 in the original version, we revised it into "A global scheme of cloud droplet parameterization" in line 168-169 in the revised version.

62: "response to aerosol particle increases"
Re: we have modified this sentence to "exhibits a sublinear relationship to aerosol number concentration (N_{CN})" in line 74-75 in the revised version.

63-64: "as is different from"
Re: we have replaced it with "this is different from" in line 77 in the revised version.

75-76: "and its characteristics like nucleation, …"
Re: we have removed that sentence and added "and the formation and growth rate of new particles may be larger than that of relatively clean atmosphere." in line 88, 89 in

the revised version.

101: “catering”
Re: we have replaced it with “cooking” in line 114 in the revised version.

103: “. And”
Re: We removed the “And” in line 116 in the revised version.

103: “the radiation in summer is relatively strong”
Re: we have corrected the sentence to “The radiation in summer is stronger than other seasons,” in line 116 in the revised version.

104-105: “contribute many CCN size particles”
Re: have been corrected as “contribute many CCN size-relevant particles” in line 118 in the revised version.

109: “) ,”
Re: Thank you for your careful checking, we removed the extra blank space between “)” and “,” in line 121.

110: “particle sizes”
Re: it has been corrected to “the size-classified particles” in line 124 in the revised version.

119-120: “measurements were deployed at ground level and at the 260 m level”
Re: we have removed “and at the 260 m level” and added some statement in lines 118-119 in the revised version, or as “The instruments during the campaign were deployed in a container at ground level (~8 m on a meteorological tower).”.

120: “2017), Before”
Re: we have revised this section and check carefully with the revised version. .

124: “Black carbon (BC), the BC”
Re: we have replaced it with “black carbon (BC), and the BC” in line 131 in the revised version.

128-129: “their surface water vapor phase equilibrium (supersaturation ratio) will gradually increase”
Re: We have rewritten this part, and removed this sentence in line 136 in the revised version.

141: “formula (5),”
Re: We have revised this section and the κ-Köhler theory refers to Eqs. 1 in line 140 in the revised version.

145: “the next calculation rule”
Re: we have replaced it with “following equation” in line 149 in the revised version.

152: “cloud parametric model”
Re: we have replaced it with “A global scheme of cloud droplet parameterization” in lines 168-169 in the revised version.

168: “(8)”
Re: we have revised this section and the equation refers to (7) in line 190 in the revised version.

174: “field observation or empirical”
Re: we have modified this sentence to “the prescribed updraft velocity used in this study is referred from previous studies” in lines 193-194 in the revised version.

Figure 1: the x-axis is shows day and not time. It looks like some vertical axis have been cut, which should be explained in the text.
Re: revised. See the Figure 2 in the revised version.

187: “As a typical NPF event…” is a long and confusing sentence
Re: the sentence has been revised as “As a typical NPF event includes the sudden appearance and continuous growth of particles smaller than 25nm, and the “banana” shape can be seen on the particle size spectrum (Dal Maso et al., 2005), here we selected 7 NPF events as cases to study and marked them in red box in Fig 1. The complete development process of these NPF events can be clearly observed. ” in lines 259-262 in the revised version.

195: “The variation of N_{CCN} seems to be more consistent with that of N_{CN} than N_d”
Re: we have corrected this sentence with “The variation trend of N_{CN} is more correlated with that of N_{CCN} than N_d (also see Fig. 4, Table S5).” in lines 270-271 in the revised version.

199: “time node”
Re: This section about the evaluating the effect of NPF to N_d have been rewritten and reorganized in lines 201-235. And we have replaced the all “time node” with “time” in lines 201-235 in the revised version.
200: “we evaluating”
Re: we have revised the section in lines 201-235, and removed this sentence “…when we evaluating the CCN...”.

273: “linear correlated”
Re: we have replaced it with “linearly related” in line 318 in the revised version.

203: “hours later after”

Re: We should replace it with “hour later than t_{start}”, and we have revised and rewritten this part in lines 201-235 in the revised version.

217: “2017, respectively”

Re: we have revised this figure in lines 236-240 and removed this sentence.“, respectively” in line 299 in the revised version.

238-240: “The rise of environmental supersaturation…”

Re: We have revised this part in 201-235 and removed it..

250: “define this impact continues”

Re: We have revised this part in lines 201-235and removed it.

276: “was thought caused”

Re: we have replaced it with “was believed to be caused by” in line 322 in the revised version.

277: “updraft velocities generate more water vapor”

Re: we have replaced this sentence with “Although the larger updraft velocities can achieve greater supersaturation in adiabatic ascending clouds and can form more N_d” in lines 323-325 in the revised version.

278 “form more Nd”

Re: Revised as “more particles can be activated into cloud droplets” in line 324 in the revised version.

279-281: “This fully suggests the difference between the fixed..”

Re: Corrected as “This is fully suggested by the difference between the calculated NCCN using the constant S and the Nd using the variable Smax in the air parcels.” in lines 325-326 in the revised version.

281: “Because in the actual environment, it is often unable…”

Re: Replaced it with “Because in the actual environment, it is often unable to achieve the sufficient supersaturation compared to the prescribed ones that are preset in the instrument.” in lines 327-328 in the revised version.

285: “The latter”

Re: we have revised it as “The S_{max}” in line 331 in the revised version.

315: “nucleation particles condense and coagulate”

Re: Corrected as “the condensation and coagulation of nucleation particles” in line 363 in the revised version.

330: “accumulated mode particles”

Re: Corrected as “accumulation mode particles” in line 372 in the revised version.

342: “due to that there”
Re: We have revised it as “, because there” in line 384 in the revised version.

343: “pre-exist”
Re: Replaced as “pre-existing” in line 385 in the revised version. And we have change all the “pre-exist” into “pre-existing” in the revise version.

353: “Chang”
Re: we have revised this figure title in lines 395-397 and removed it..

354: “get the this proportion”
Re: Revised this figure title in lines 395-397 and removed it.
363: “demonstrated related”
Re: Replaced with “and accumulation mode (~100-120 nm) according to Brines et al ” in line 427 in the revised version.

364: “Brines, M et al.”
Re: We have corrected it as “Brines et al. (2015)” in line 427 in the revised version.

364: “primarily”
Re: We have modified this part in lines 419-439 and removed it. with “Those particles from primary emissions” in lines 454-455 in the revised version.

367: “after one hour of the eruption of newly formed”
Re: We have replaced it with “one hour after the burst of newly formed particles” in lines 405 in the revised version.

376: “Mass fraction of aerosol composition”
Re: Replaced as “mass fraction of aerosol chemical components” in lines 442-443 in the revised version.

395: “taking as a background”
Re: we have replaced the total sentence with “Then the increment of N_{CCN} and N_d from NPF are obtained from the PNSD of NPF mode, and the increment of N_{CCN} and N_d from primary emissions are obtained by subtracting the increment of N_{CCN} and N_d by NPF from the total increment of N_d. ” in lines 436-439 in the revised version.

409: “CCN and cloud droplet.”
Re: Revised as “N_{CCN} and N_d” in line 469 in the revised version.

414: “V that can provide more sufficient water vapor”
Re: We have replaced the sentence with “The effect of water vapor competition

becomes smaller at larger V at which the greater S_{max} can be achieved." in lines 474-475 in the revised version.

418: "As a result, although 418 a larger increase…"
Re: we have revised it as "As a result, although a larger enhancement" in line 479 in the revised version.

425: "variance of CDNC"
Re: Corrected as "variation of CDNC" in line 486 in the revised version.

427: "leading to ~50% enhancement in the year from1850 to 2000 change in cloud albedo."
Re: Revised as "leading to about 50% enhancement in the year from1850 to 2000 change in cloud albedo" in lines 488 in the revised version.

429: "related cloud microphysical progress research"
Re: we have replaced it with "the research of the microphysical process of aerosol-cloud" in lines 490-491 in the revised version.

433, 436: Code/data availability and Competing interests should be separate sections
Re: Corrected.

504: "droplets[J]. Transactions"
Re: Corrected as "droplets, Transactions" in line 585 in the revised version.

516: "Zhang F*., Li"
Re: Corrected as "Li Y., Zhang F., " in line 597 in the revised version.

613: "Zhang, R., … Li"
Re: Corrected as "Zhang, R., Li, Z." in line 718 in the revised version.

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 (ΔN_{CCN} or ΔN_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_{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_{start}.

Since there need 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 $t_{start,CCN}$ and $t_{end,CCN}$ respectively, the time series of N_{CCN} was firstly divided by the N_{CCN} at t_{start} at each prescribed supersaturation, to derive the normalized time series of N_{CCN}, denoted as R_s. The equation is written as follows,

$$R_S = \frac{CCN_S}{CCN_{S,t_{start}}} \quad (8)$$

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)."

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% 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 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_{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_{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)…."

Kalkavouras, P. , Bougiatioti, A. , Kalivitis, N. , Stavroulas, I. , and Mihalopoulos, N.: Regional new particle formation as modulators of cloud condensation nuclei and cloud droplet number in the eastern mediterranean. Atmos. Chem. Phys., 19(9), 6185-6203, doi: 10.5194/acp-19-6185-2019,2019.

Peng, J. F., Hu, M., Wang, Z. B., Huang, X. F., Kumar, P., Wu, Z. J., Guo, S., Yue, D. L., Shang, D. J., Zheng, Z., and He, L. Y.: Submicron aerosols at thirteen diversified sites in China: size distribution, new particle formation and corresponding contribution to cloud condensation nuclei production, Atmos. Chem. Phys., 14, 10249–10265, doi:10.5194/acp-14-10249-2014, 2014.

Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements, Atmos. Chem. Phys., 15, 13071–13083, doi:10.5194/acp-15-13071-2015, 2015.

Ma, N., Zhao, C., Tao, J., Wu, Z., Kecorius, S., Wang, Z., Größ, J., Liu, H., Bian, Y., Kuang, Y., Teich, M., Spindler, G., Müller, K., van Pinxteren, D., Herrmann, H., Hu, M., and Wiedensohler, A.: Variation of CCN activity during new particle formation events in the North China Plain, Atmos. Chem. Phys., 16, 8593–8607, doi:10.5194/acp-16-8593-2016, 2016.

Ren, J., Zhang, F., Wang, Y., Collins, D., Fan, X., Jin, X., Xu, W., Sun, Y., Cribb, M., and Li, Z.: Using different assumptions of aerosol mixing state and chemical composition to predict CCN concentrations based on field measurements in urban Beijing, Atmos. Chem. Phys., 18, 6907–6921, doi:10.5194/acp-18-6907-2018, 2018.

Zhang, F., Ren, J., Fan, T., Chen, L., Xu, W., Sun, Y.: Significantly enhanced aerosol CCN activity and number concentrations by nucleation‐initiated haze events: A case study in urban Beijing. J. Geophys. Res.-Atmos., 124. doi:10.1029/ 2019JD031457, 2019

Fan, X., Liu, J., Zhang, F., Chen, L., Collins, D., Xu, W., Jin, X., Ren, J., Wang, Y., Wu, H., Li, S., Sun, Y., and Li, Z.: Contrasting size-resolved hygroscopicity of fine particles derived by HTDMA and HR-ToF-AMS

measurements between summer and winter in Beijing: the impacts of aerosol aging and local emissions, Atmos. Chem. Phys., 20, 915–929, https://doi.org/10.5194/acp-20-915-2020, 2020.

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

V or S	D_c	$\triangle N_{d\_NPF}$ or ΔN_{CCN_NPF}		$\triangle N_{d\_PE}$ [a] or ΔN_{CCN_PF} [a]		$\triangle N_{d\_total}$ or $\triangle N_{CCN\_total}$
$m\ s^{-1}$,or %	nm	cm^{-3},	%	cm^{-3},	%	cm^{-3}
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}						
0.2%	109	654	92.0%	57	8.0%	711
0.4%	69	1356	87.2%	199	12.8%	1555
0.60%	52	1680	87.10%	249	12.90%	1929
0.80%	43	1801	85.00%	318	15.00%	2119

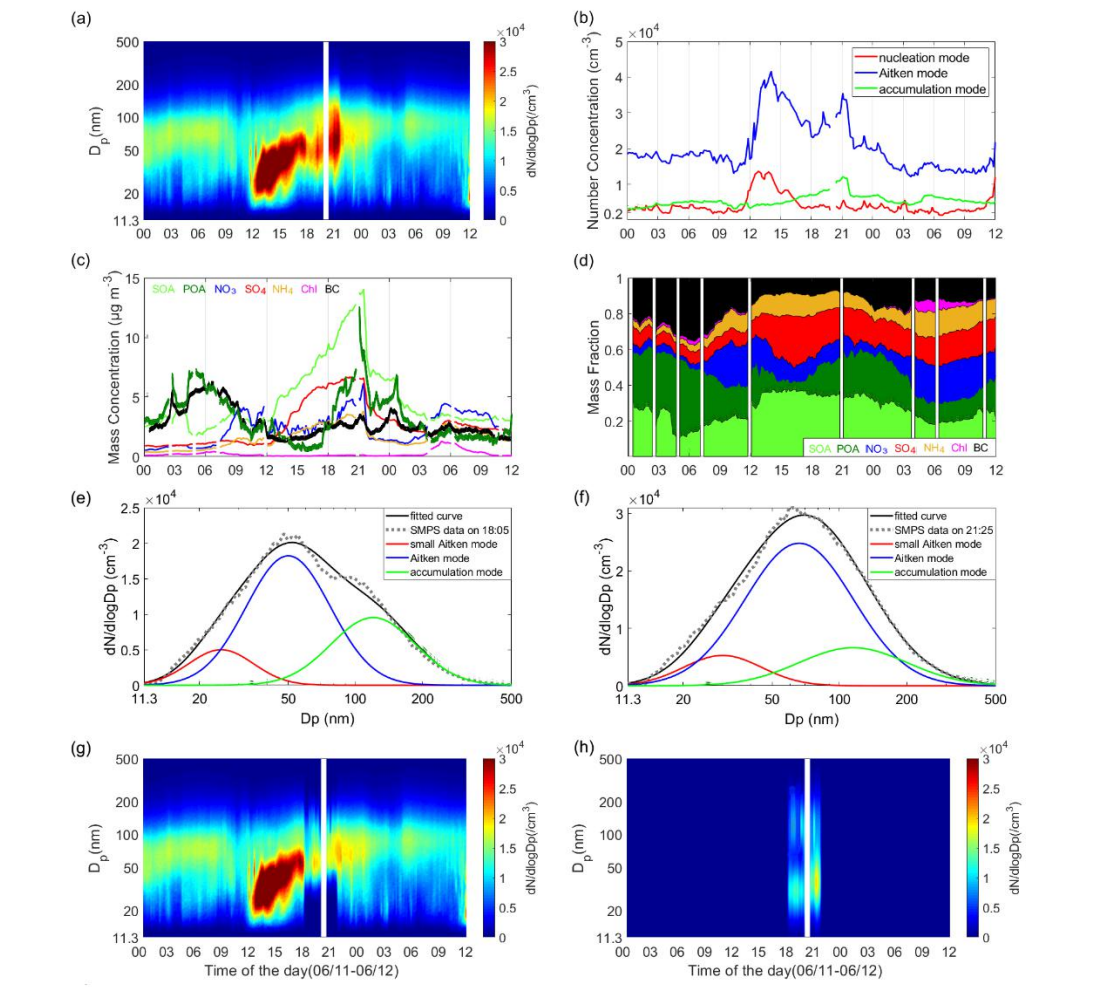

Figure 8. Diurnal variations of the (a) aerosol size distribution, (b) particle number concentrations for different size modes, (c) mass concentrations of aerosol chemical composition, and (d) mass fraction of aerosol chemical components, (e) (f) fitted three modes of the particle number concentration 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; Altstädter, 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…."

Kerminen, V. M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi, E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala, M., and Petäjä, T.: Cloud condensation nuclei production associated with atmospheric nucleation: a synthesis based on existing literature and new results, Atmos. Chem. Phys., 12, 12037-12059, doi:10.5194/acp-12-12037-2012, 2012.

Altstädter, B., Platis, A., Wehner, B., Scholtz, A., Wildmann, N., Hermann, M., Käthner, R., Baars, H., Bange, J., and Lampert, A. (2015). ALADINA – an unmanned research aircraft for observing vertical and horizontal distributions of ultrafine particles within the atmospheric boundary layer. Atmos. Meas. Tech., 8, 1627–1639, doi:10.5194/amt-8-1627-2015.

Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu, B. (2017). Aerosols and boundary-layer interactions and impact on air quality. Natl. Sci. Rev., 4, 810–833, https://doi.org/10.1093/nsr/nwx117, 2017.

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 $n^s(s')$) and total number of particles with S_c smaller than S, $F^S(S)$, which is given by

$$F^S(S_x) = \int_0^{S_x} n^S(S')dS' \quad (6)$$

Where the S_x 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 $F^S(S_x)$ can be calculated by the integration of $n^S(S')$ from the lower limit 0 to upper limit S_x…."

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 (ΔN_{CCN} or ΔN_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_{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_{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 $t_{start,CCN}$ and $t_{end,CCN}$ respectively, the time series of N_{CCN} was firstly divided by the N_{CCN} at t_{start} at each prescribed supersaturation, to derive the normalized time series of N_{CCN}, denoted as R_s. The equation is written as follows,

$$R_S = \frac{CCN_S}{CCN_{S,t_{start}}} \quad (8)$$

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)…."

Kalkavouras, P. , Bougiatioti, A. , Kalivitis, N. , Stavroulas, I. , and Mihalopoulos, N.: Regional new particle formation as modulators of cloud condensation nuclei and cloud droplet number in the eastern mediterranean. Atmos. Chem. Phys., 19(9), 6185-6203, https://doi.org/10.5194/acp-19-6185-2019,2019.

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Wu, Z. J., Poulain, L., Birmili, W., Größ, J., Niedermeier, N., Wang, Z. B., Herrmann, H., and Wiedensohler, A.: Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements, Atmos. Chem. Phys., 15, 13071–13083, doi:10.5194/acp-15-13071-2015, 2015.

Ma, N., Zhao, C., Tao, J., Wu, Z., Kecorius, S., Wang, Z., Größ, J., Liu, H., Bian, Y., Kuang, Y., Teich, M., Spindler, G., Müller, K., van Pinxteren, D., Herrmann, H., Hu, M., and Wiedensohler, A.: Variation of CCN activity during new particle formation events in the North China Plain, Atmos. Chem. Phys., 16, 8593–8607, doi:10.5194/acp-16-8593-2016, 2016.

Ren, J., Zhang, F., Wang, Y., Collins, D., Fan, X., Jin, X., Xu, W., Sun, Y., Cribb, M., and Li, Z.: Using different assumptions of aerosol mixing state and chemical composition to predict CCN concentrations based on field measurements in urban Beijing, Atmos. Chem. Phys., 18, 6907–6921, doi:10.5194/acp-18-6907-2018, 2018.

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Fan, X., Liu, J., Zhang, F., Chen, L., Collins, D., Xu, W., Jin, X., Ren, J., Wang, Y., Wu, H., Li, S., Sun, Y., and Li, Z.: Contrasting size-resolved hygroscopicity of fine particles derived by HTDMA and HR-ToF-AMS measurements between summer and winter in Beijing: the impacts of aerosol aging and local emissions, Atmos. Chem. Phys., 20, 915–929, https://doi.org/10.5194/acp-20-915-2020, 2020.

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_{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-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_{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 levelon 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 ΔN_{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 ΔN_{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 NCCN", 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}."

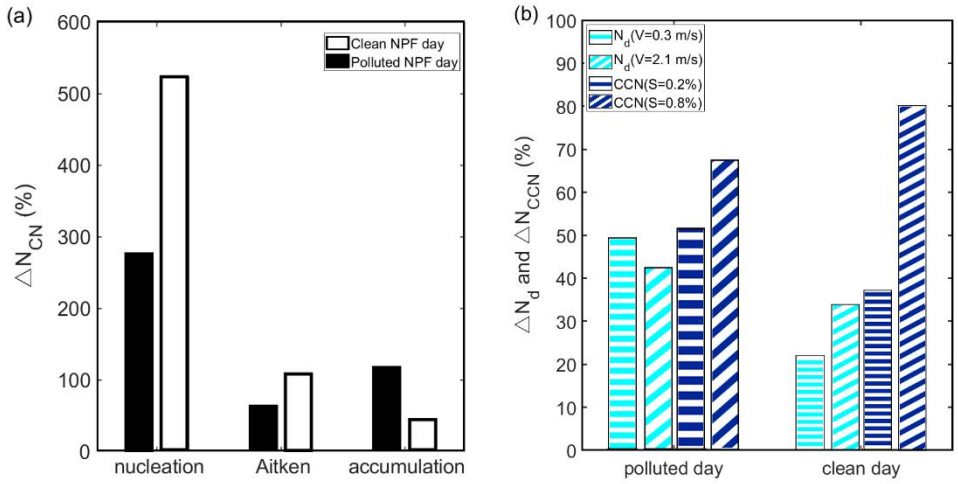

Figure 7. Comparison of the increments of (a) total particle number concentration (N_{CN}), and (b) CCN number concentration (N_{CCN}) and cloud dropet number concentration (N_d) between the two different typical NPF events.

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.