

Reviewer #2:

In the manuscript “Secondary aerosol formation alters CCN activity in the North China Plain”, the authors conducted a field study in North China Plain and investigated the influence of second aerosol (SA) formation on CCN activity and on the calculated CCN number concentrations derived from particle number size distribution (PNSD). The CCN activity at 0.05% supersaturation (SS) was discussed. The authors focused on CCN activation at low SS where mainly accumulation mode particles act as CCN and thus on cases of SA in the presence of accumulation mode particles. They found that at two different RH, SA formation had different influence on CCN activity of aerosols. At high RH (minimum RH>50%), SA mass mostly added to larger particles (>300 nm), which resulted in weaker enhancement of CCN activity for per SA mass added as these larger particles were already CCN-active before SA formation. At low RH (minimum RH<30%), SA mass mostly added to smaller particles (<300 nm), which resulted in stronger enhancement of CCN activity for per SA mass added as smaller particles were not CCN-active before SA formation. In addition, they parameterized maximum activation fraction (MAF) using the correlation of MAF with hygroscopic particle number fraction or with mass fraction of SA. The calculated CCN concentrations derived from PNSD using parameterized MAF, campaign average activation diameter and width of activation curve matched better with measured ones compared with using PNSD and kappa from either chemical composition or hygroscopic growth.

How aerosol formation and growth affect CCN activity is an important question. The manuscript provides valuable case study on how secondary aerosol formation influence CCN activity for low stratus clouds and fogs. This study carried out comprehensive measurement of aerosol related to CCN activity/hygroscopicity. The findings are interesting. However, I have some comments about the manuscript to address before it is considered for publishing in ACP.

Response: Thanks for your comments. Suggestions and comments are addressed point-by-point and corresponding responses are listed below.

Major comments:

1. The manuscript only discussed the results at 0.05% SS. How do the findings depend on SS? What about the results at other SS such as 0.1% and 0.2% SS, which is also typical for low stratus clouds? In addition, I suggest explicitly specifying SS when CCN activity or CCN concentration is discussed.

Response: Thanks for your suggestion. We have added the variations of CCN number concentration at the five measured SSs into Fig. 1, the variations of SPAR and the ratios between CCN number concentration and PM_{2.5} at SS of 0.07% and 0.2% in Fig. S1 and the diurnal variations of SPAR at SS of 0.07% and 0.2% in Fig. S2, as follow:

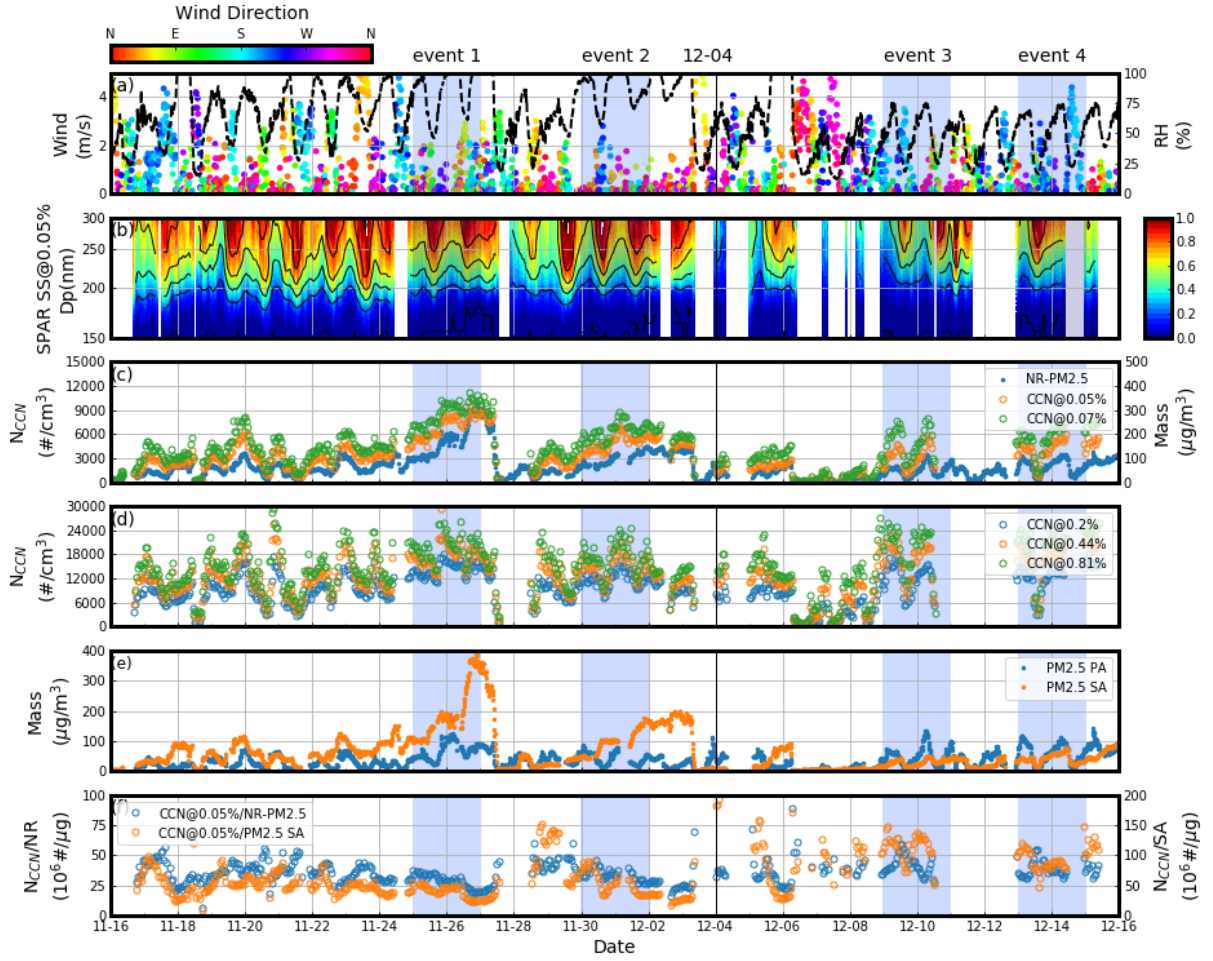


Fig 1. Overview of the measurements during the campaign: (a) dots represent wind speed with color indicating wind direction, and black lines represent RH; (b) SPAR under SS of 0.05%; (c) blue, green and yellow dots represent NCCN under SS of 0.05% and 0.07%, and mass concentration of NR-PM_{2.5}, respectively; (d) blue, green and yellow dots represent NCCN under SS of 0.2%, 0.44% and 0.81%, respectively; (e) blue and yellow dots represent mass concentration of PM_{2.5} PA and PM_{2.5} SA respectively; (f) blue and yellow dots represent ratio between NCCN under SS of 0.05% and mass concentration of NR-PM_{2.5} and PM_{2.5} SA, respectively. There were four events with significant enhancements of NCCN during the blue shaded periods.

As the Fig. 1 shows, the variations of NCCN at 0.07% were similar to those at 0.05%, which follow the variations of SA mass concentration, while the variations of NCCN at higher SSs including 0.4% were different from the variations of SA mass concentration, especially under high RH conditions, suggesting different responses to SA formation. We have added these discussions into the first paragraph of section 3.1 as follow:

“It should be noted that variations of N_{CCN} at 0.07% were similar to those at 0.05%, which followed the variations of SA mass concentration. While at higher SSs, the variations of N_{CCN} differed from those of SA mass concentration, especially under high RH conditions, suggesting different responses of CCN activity towards distinct SA formation processes.”

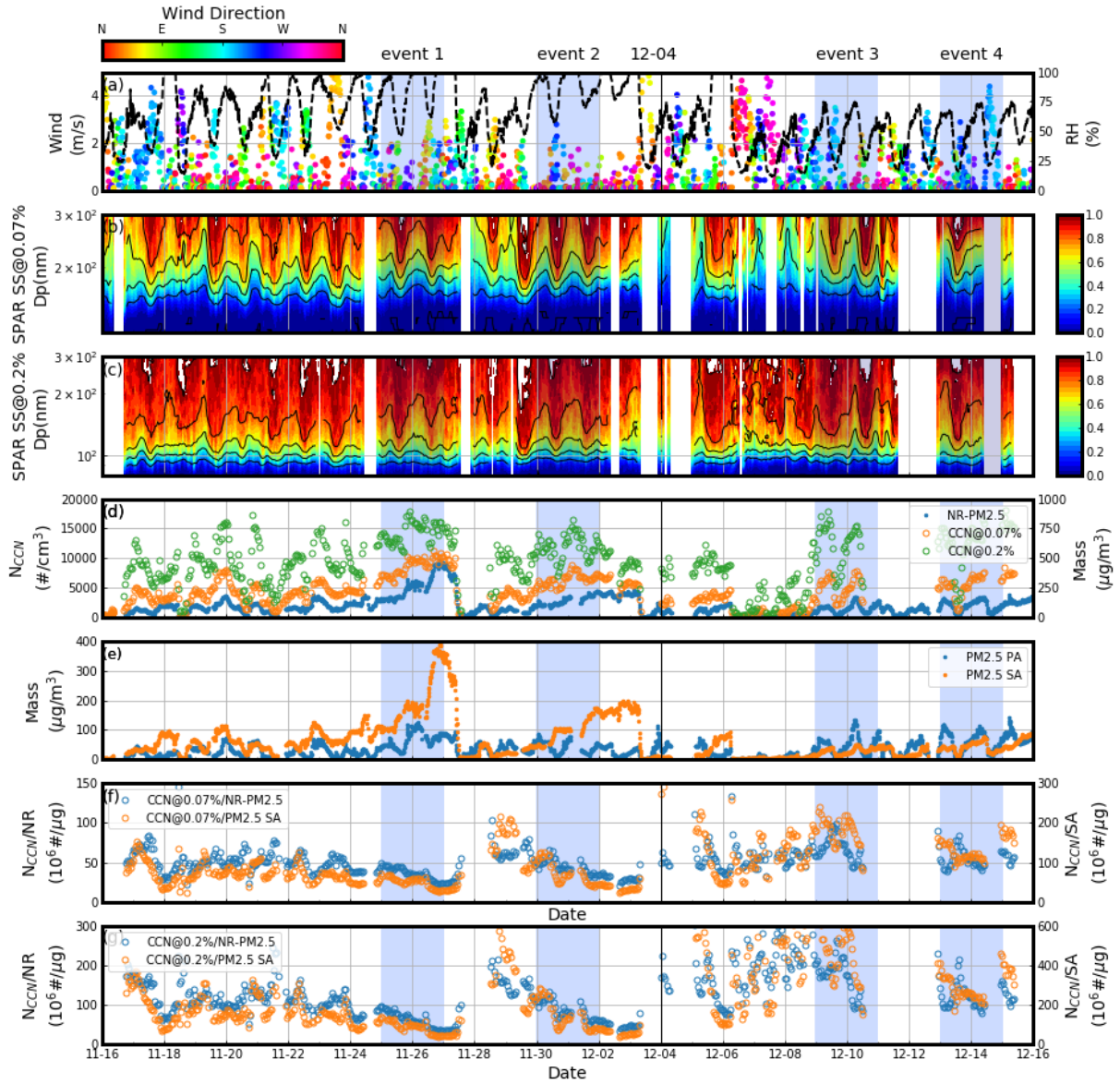


Fig S1. Overview of the measurements during the campaign: (a) dots represent wind speed with color indicating wind direction, and black lines represent RH; (b) SPAR under SS of 0.07%; (c) SPAR under SS of 0.2%; (d) blue, green and yellow dots represent NCCN under SS of 0.07% and 0.2%, and mass concentration of NR-PM2.5, respectively; (e) blue and yellow dots represent mass concentration of PM2.5 PA and PM2.5 SA respectively; (f) blue and yellow dots represent ratio between NCCN at SS of 0.07% and mass concentration of NR-PM2.5 and PM2.5 SA, respectively. (g) blue and yellow dots represent ratio between NCCN at SS of 0.2% and mass concentration of NR-PM2.5 and PM2.5 SA, respectively. There were four events with significant enhancements of NCCN during the blue shaded periods.

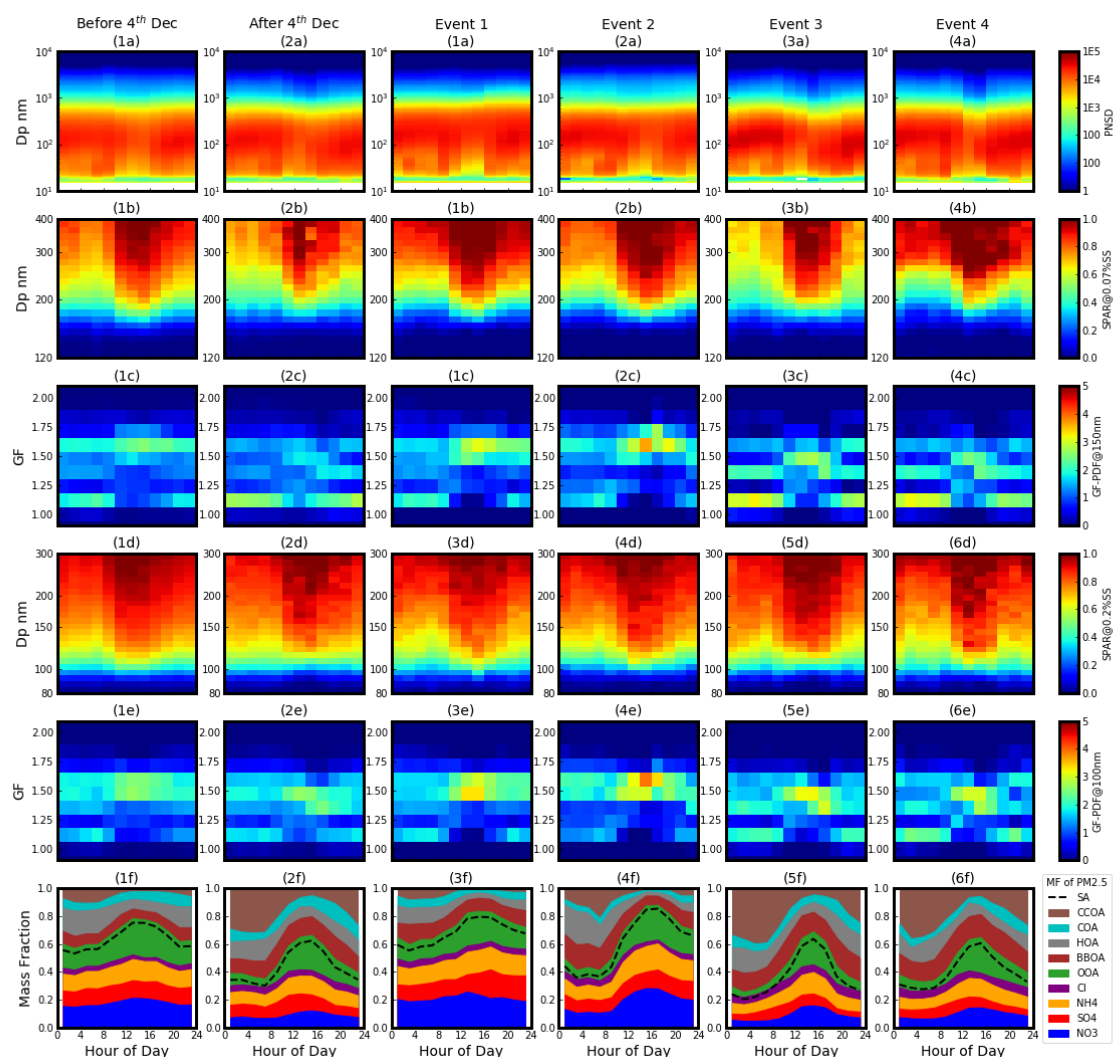


Fig S2. Diurnal variation of (a) PNSD, (b) SPAR at SS of 0.07%, (c) GF-PDF at 150nm, (d) SPAR at SS of 0.2%, (e) GF-PDF at 100nm and (f) mass fraction of different PM_{2.5} chemical species during high RH periods before 4th Dec (1) low RH periods after 4th Dec (2) and the four events (3-6), including OA factors: hydrocarbon-like OA (HOA), cooking OA (COA), biomass burning OA (BBOA), coal combustion OA (CCOA), and oxygenated OA (OOA).

As shown in Figs. S1 and S2, the variations of SPAR and NCCN/PM at SS of 0.07% are similar but lighter, compared with those at SS of 0.05%. For SS of 0.2%, the difference of SPAR between different periods or events are smaller (Fig. S1), and so did the diurnal variations of SPAR and GF-PDF at particle size of 100 nm (Fig. S2). While for SS of 0.2%, the difference of SPAR between different periods or events are smaller (Fig. S1), and so did the diurnal variations of SPAR and GF-PDF at particle size of 100nm (Fig. S2). Because CCN activity at SS of 0.2% was strong enough (indicated by SPAR value close to 1) in particle size range where the SA formation dominates, and thus the different SA formations under high or low RH conditions cannot lead to significant variations of CCN activity at SS of 0.2%. In summary, based on CCN measurements in this study, the RH-dependent influence of SA formation on CCN activity can be found obviously at SSs of 0.05% and 0.07%. As the variations of CCN activity at SS of 0.07% were quite similar to those at SS of 0.05,

further analysis was only based on CCN activity at SS of 0.05%. We have added a paragraph of these discussions after the first paragraph of section 3.2 (discussing Fig. 2) as follow:

“Besides SS of 0.05%, variations of SPAR at SSs of 0.07% and 0.2% are also shown in Figs. S1 and S2 in the supplement. And as shown in Figs. S1 and S2, the variations of SPAR and NCCN/PM at SS of 0.07% are similar but lighter, compared with those at SS of 0.05%. While for SS of 0.2%, the difference of SPAR between different periods or events are smaller (Fig. S1), and so did the diurnal variations of SPAR and GF-PDF at particle size of 100nm (Fig. S2). Because CCN activity at SS of 0.2% was strong enough (indicated by SPAR value close to 1) in particle size range where the SA formation dominates, and thus the different SA formations under high or low RH conditions cannot lead to significant variations of CCN activity at SS of 0.2%. In summary, based on CCN measurements in this study, the RH-dependent influence of SA formation on CCN activity can be found obviously at SSs of 0.05% and 0.07%. As the variations of CCN activity at SS of 0.07% were quite similar to those at SS of 0.05, further analysis was only based on CCN activity at SS of 0.05%.”

In addition, we have also added the specification of the SS where CCN activity and CCN number concentration are discussed in the manuscript.

2. I was somewhat surprised to notice that MAF only reached 0.4-0.6 in Fig. 3. Why were the data larger than 300 nm excluded (L148)? Did the activation fraction reach around one at larger sizes? If one fit Eq.7 to e.g. blue curves in Fig. 3a only till 300 nm, the D_a derived at half MAF might be incorrect.

Response: Thanks for your comments. The low value of MAF was mainly due to the limited range of measured particle size and the large fraction of POA with low hygroscopicity which can be seen in the measurement of particle chemical compositions. The reason of excluding the data for larger than 300 nm is that there is higher noise in CCN measurement due to the very low particle number concentration in this size range. To evaluate the influence of the size cut-off, we have expanded the upper size limit of SPAR to about 400 nm and obtained new fitting parameters as shown in Fig. R1. Compared with the original parameters, new MAF and D_a are both higher, especially at SSs of 0.05%. We have also applied these new values of fitting parameters into our study and revised the manuscript accordingly, as described below.

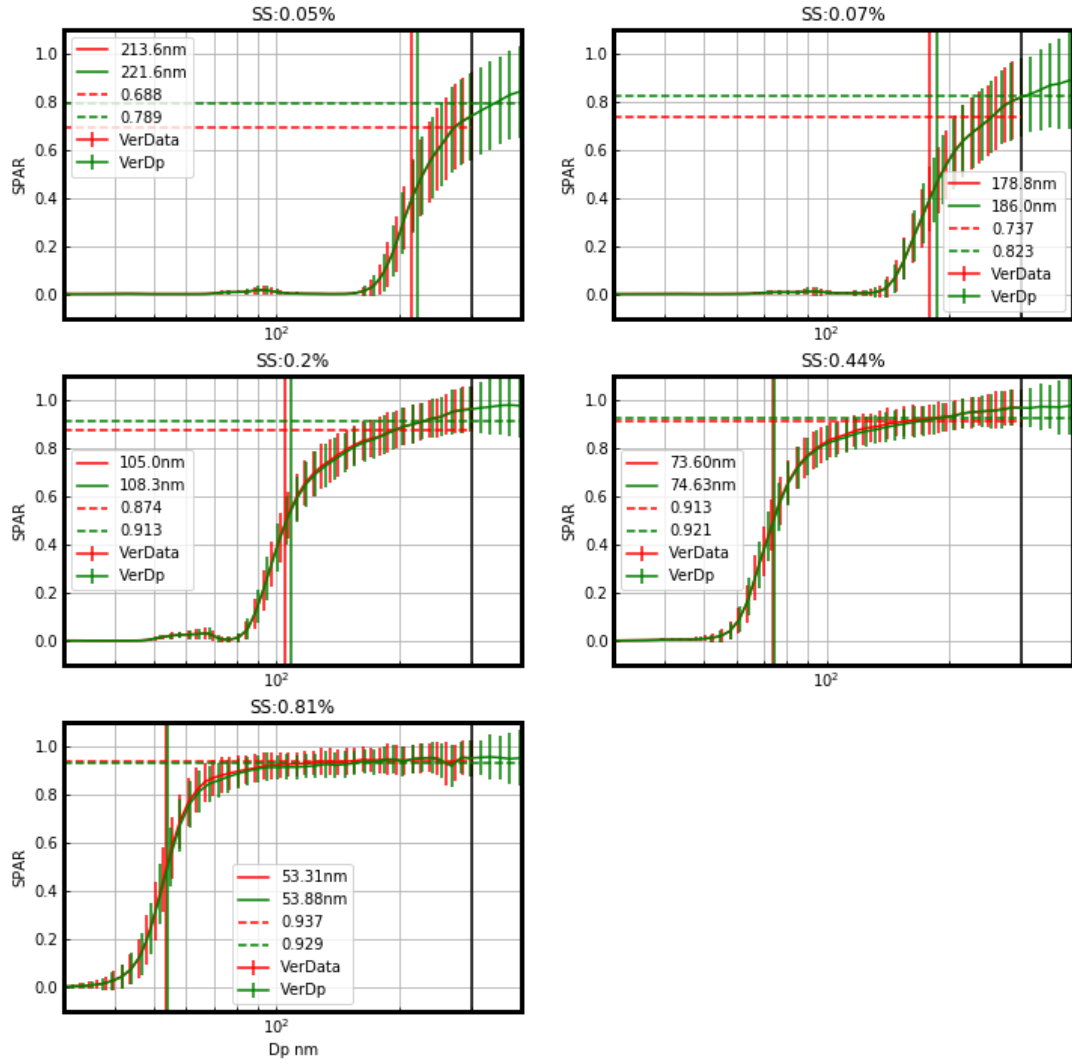


Fig. R1. SPAR and the corresponding fitting parameters of MAF and D_a for the original (red) and the expanded particle size ranges (green) at the five measured SSs. The vertical red and green lines indicate the original D_a and the new D_a , respectively. The vertical black line indicates the particle size of 300nm.

Furthermore, we have revised the corresponding parts related to SPAR fitting parameters, including Figs. 3, 5, 6 and 7 (shown below). In the particle size range larger than 300 nm, the SPAR is still lower than 1 at SS of 0.05% (Fig. 3). This is because for particle size of ~ 390 nm, kappa value higher than 0.1 is needed for CCN activation at SS of 0.05%. As the temporal variations of SPAR fitting parameters stay the same, the conclusions based on Fig. 3 are still valid. In the updated Fig. 5, diurnal variations of the ratios between calculated NCCN and measured NCCN are stronger and the standard deviations are higher. These strong diurnal variations and larger deviations are because both the fitting parameters of MAF and their difference from the campaign averaged MAF become larger. In Figs. 6 and 7, there are difference of MAR_SPAR and the corresponding calculated NCCN (based on MF_{SA} and NF_{hygro}) by expanding the size range of SPAR. As the Figs. 6c and 7c show, the calculated NCCN become lower, which is mainly due to the higher values of new D_a shown in Fig. R1. Thus, compared with the original results, correlations in Figs. 6b, 6c, 7b and 7c are nearly the same except

that the slopes decrease by about 0.1. Nevertheless, as in particle size range larger than 400nm, the PNSDs are low and the resultant influence on NCCN are small, the conclusions in Figs. 5, 6 and 7 are still valid.

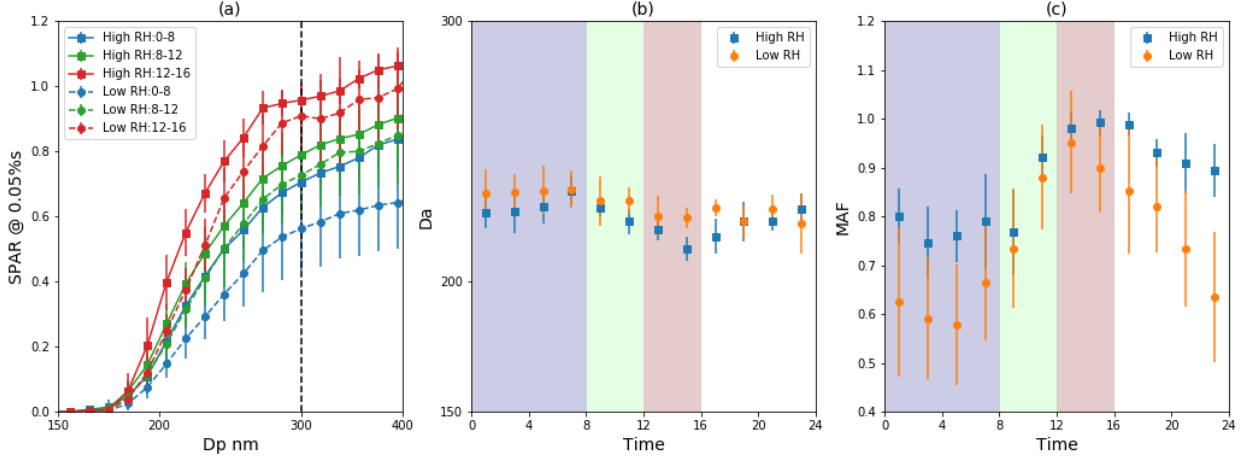


Fig 3. (a) The averages of SPAR curves at SS of 0.05% in three different time periods (blue: 0:00-8:00; green: 8:00-12:00; red: 12:00-16:00) during high (squares with solid line) and low (dots with dashed line) RH events. Diurnal variation of (b) Da and (c) MAF under high (blue) and low (yellow) RH conditions. The blue, green and red shades correspond to with the three periods in (a & d). Error bars indicate the standard deviations of data.

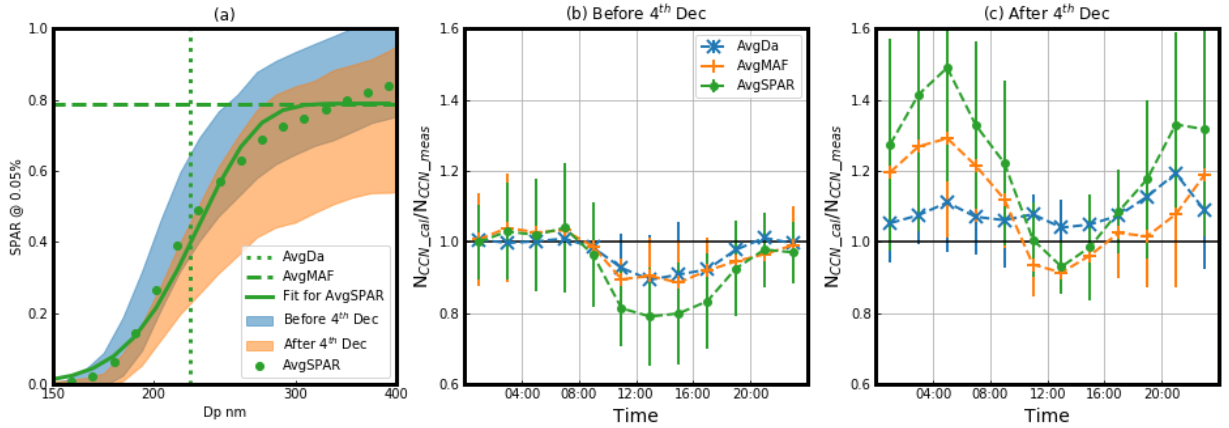


Fig 5. (a) The averaged SPAR during the campaign (green scatters), the corresponding fitting curve (green line) and the averaged fitting parameters (dotted line for Da and dashed line for MAF). The blue and yellow shaded areas represent the variations of SPAR before 12-04 and after 12-04, respectively. The ratio between calculated NCCN and measured NCCN under (b) before 12-04 and (c) after 12-04. Bars represent one standard deviation and colors represent different calculation of SPAR curves: green represent average SPAR during the campaign (AvgSPAR), yellow represent SPAR calculated with average Da and real-time MAF (AvgDa) and blue represent SPAR calculated with average MAF and real-time Da (AvgMAF).

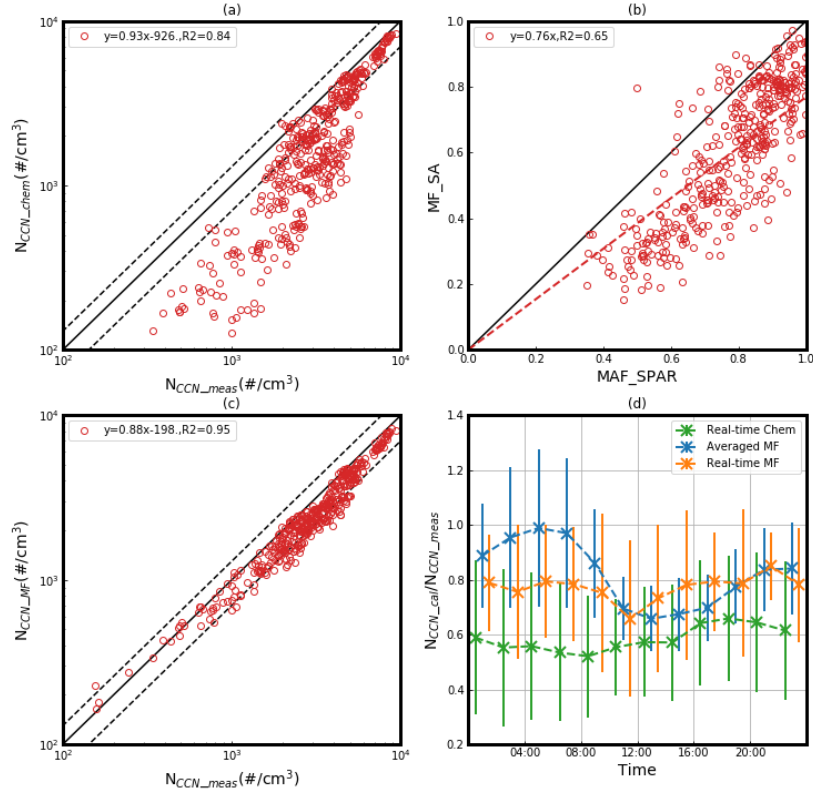


Fig 6. (a) The comparison between calculated N_{CCN} based on kappa derived from bulk particle chemical compositions (N_{CCN_chem}) and measured N_{CCN} at SS of 0.05%. (b) The correlation between MAF and mass fraction of secondary aerosol (MF_{SA}). (c) the comparison between calculated N_{CCN} based on SPAR derived from real-time MFSA and average Da (N_{CCN_MF}) and measured N_{CCN} . The black dashed lines represent the relative deviation of 30%. (d) the diurnal variations of the ratio between the calculated and measured N_{CCN} during the whole campaign based on different methods (green: N_{CCN_chem} ; blue: N_{CCN} calculated based on SPAR derived from averaged MF_{SA} and average Da ; yellow: N_{CCN_MF}).

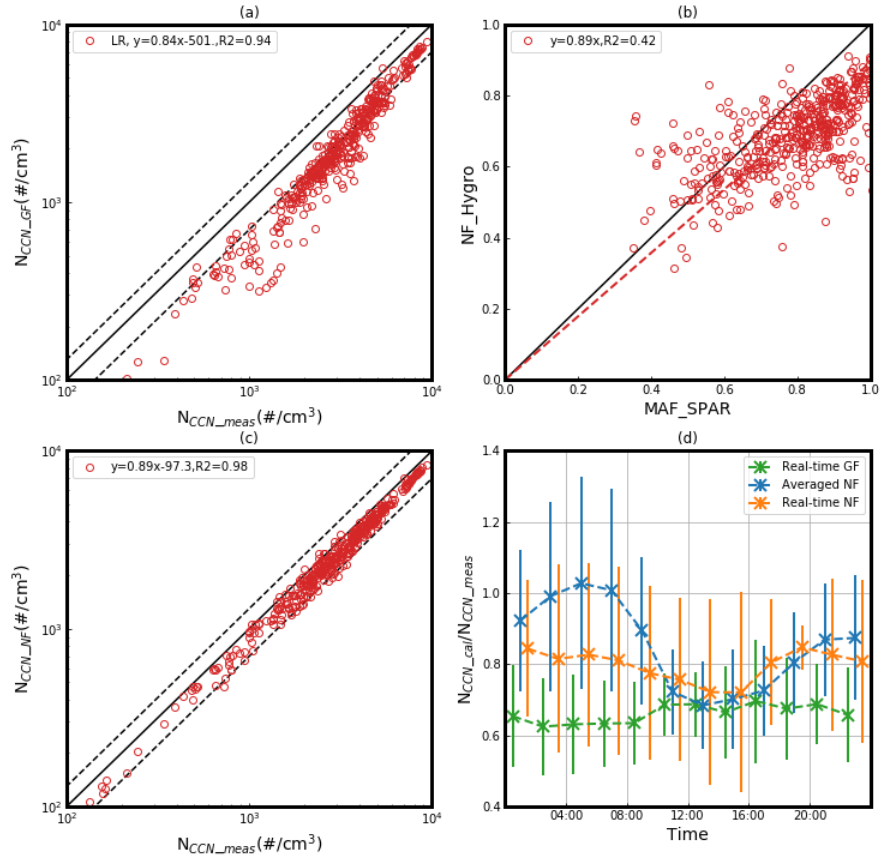


Fig 7. (a) The comparison between calculated NCCN based on kappa derived from bulk GF at 200 nm (NCCN_GF) and measured NCCN at SS of 0.05%. (b) The correlation between MAF and number fraction of hygroscopic particles (NFhygro, GF>1.2). (c) The comparison between calculated NCCN based on SPAR derived from real-time NFhygro and average Da (NCCN_NF) and measured NCCN. The black dashed lines represent the relative deviation of 30%. (d) the diurnal variations of the ratio between the calculated and measured NCCN during the whole campaign based on different methods (green: NCCN_GF; blue: NCCN calculated based on SPAR derived from averaged NFhygro and average Da; yellow: NCCN_NF).

3. The authors reported two cases at high RH and only one case at low RH. It would be helpful to discuss how general these conclusions are regarding the influence of SA on CCN activity. The authors seem to indicate that RH is the dominant factor. What about other conditions? For example, how would the size and chemical composition of existing particle affect the conclusion here?

Response: Thanks for your comments.

We agree that it's important to convince the different responses of CCN activity to different SA formations. In the revised discussions of Fig. 2, the averaged variations of CCN activity during high or low RH conditions are analyzed in front of the analyses of specific events. And as show in revised

Figs. 2(1a-1d) and 2(2a-2d) (shown below), different variations of SPAR to SA formations can be found during periods with different RH conditions. The variations of SPAR, GF-PDF and mass fraction of particle chemical compositions during the periods of high (or low) RH conditions were similar but less significant, as those during high-RH events 1 and 2 (or low-RH events 3 and 4). The four specific events (adding the 14th Dec as an events under low RH conditions) with significant variations of CCN activity during SA formation are analyzed as examples (Figs. 2(3x) to 2(6x)). In addition, we have also revised Figs. 1, 3 and 4 accordingly, and the corresponding results in these figures are still valid. We have added corresponding discussion into the first paragraph of section 3.2 as follow:

“The diurnal averages of PNSD, SPAR at SS of 0.05%, GF-PDF for 200 nm particle and mass fraction of particle chemical compositions during high RH periods before 4th Dec, low RH periods after 4th Dec and the four events are shown in Fig. 2, respectively. To be noted, ... CCN behavior. As can be seen in Figs. 2 (1b) and (2b), different variations of SPAR due to SA formations can be found during the periods with different RH conditions. The average diurnal variations of these parameters for the entire high RH stage and low RH stage as shown in Figs. 2 (1a-1d) and (2a-2d) revealed similar but more smoothed variations as in the four selected events. The four events are discussed and intercompared in the following to magnify the differences under distinct RH conditions.”

Furthermore, in this study, the main point is that different SA formations during high RH and low RH environments are responsible for the variations of CCN activity. The “high (or low) RH events” is used to refer to the SA formation events under high (or low) RH conditions for convience. As reported by Kuang et al., (2020), SA formation mechanisms and the corresponding influence on PNSD and particle chemical compositions are different during periods with different RH conditions. Thus, we investigated the variations of CCN activity measured during the same campaign and found that different SA formations can largely influence CCN activity due to variations of PNSD and particle chemical compositions. The misleading statements in the manuscript have been revised accordingly:

1. After the first sentence in Sec. 3.2. (discussing the Fig. 2), a description has been added as *“To be noted, the “high (or low) RH events” is used to refer to the SA formation events under high (or low) RH conditions for convience, and it doesn’t mean that RH caused variations of CCN behavior.”*

2. The first sentence of the second paragraph in Sec. 3.2 (discussing the Fig. 3a) has been revised as *“In Figs. 3a,d, detailed comparison of particle CCN activity during SA formation events of NCCN enhancements under different RH conditions are shown as the variations of SPAR curves.”*

3. The second sentence of the second paragraph in Sec. 3.3 (discussing the Fig. 5) has been revised as *“In former discussions, CCN activity (indicated by SPAR) revealed significant diurnal variations during this campaign, which were different during SA formations under distinct RH conditions.”*

4. The first sentence of the last paragraph in Sec. 3.3 (the summary of this section) has been revised as *“In summary, MAF exhibited strong diurnal variation that varied under different RH conditions due to different SA formation mechanisms, which ...”*

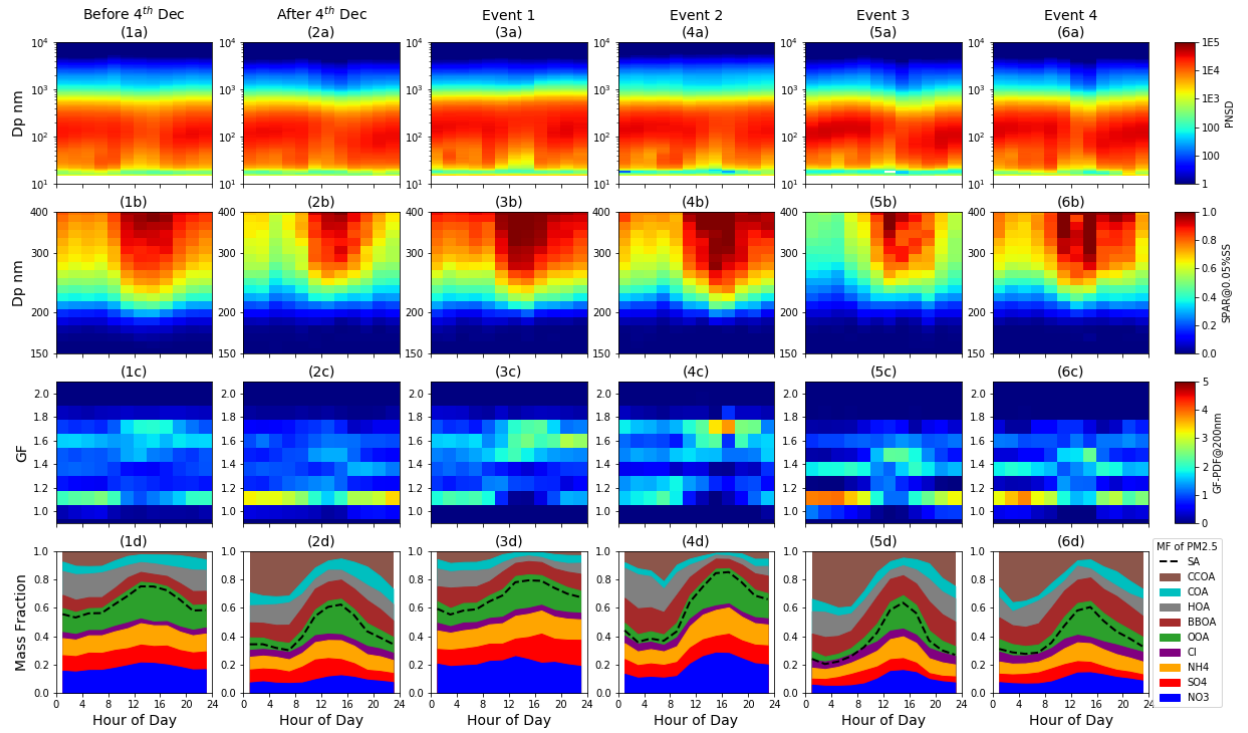


Fig 2. Diurnal variation of (a) PNSD, (b) SPAR at SS of 0.05%, (c) GF-PDF at 200nm and (d) mass fraction of different PM_{2.5} chemical species during high RH periods before 4th Dec (1), low RH periods after 4th Dec (2) and the four events (3-6), including OA factors: hydrocarbon-like OA (HOA), cooking OA (COA), biomass burning OA (BBOA), coal combustion OA (CCOA), and oxygenated OA (OOA).

4. I had some difficult time reading the manuscript. I suggest the authors streamlining the writing substantially. Additionally, there are numerous language problems. For example, in many cases, a space was missing before a unit. More specific problems are listed below.

Response: Thanks for your suggestions. We have streamlining the writing substantially and fixed language problems. These specific problems are addressed point-by-point below.

Specific comments

1. L254, it is half MAF that can represent the number fraction of CCNs to total particles at particle size around D_a . Also “represents” should be “represent”.

Response: Thanks for your comment. We have revised them accordingly.

2. L264, how are PA and SA characterized?

Response: During the campaign, PM_{2.5} PA were generally lower than 100 $\mu\text{g}/\text{cm}^3$ under both high and low RH periods. Meanwhile, PM_{2.5} SA can approach about 400 $\mu\text{g}/\text{cm}^3$, especially during the strong SA formation events under high RH conditions, but can be lower than 100 $\mu\text{g}/\text{cm}^3$ under low

RH conditions. We have added these information into the manuscript as “During the campaign, $PM_{2.5}$ PA were generally lower than $100 \mu\text{g m}^{-3}$ under both high and low RH periods. Meanwhile, $PM_{2.5}$ SA can approach about $400 \mu\text{g m}^{-3}$, especially during the strong SA formation events under high RH conditions, but can be lower than $100 \mu\text{g m}^{-3}$ under low RH conditions.”.

3. L276, how are the time ranges of these events defined? By $PM_{2.5}$ concentration?

Response: These events were chosen based on not only the RH but also the enhancement of SA. During event 3, the wind speed was generally low, the RH followed a general diurnal variations and SA mass grew steadily and continuously. Thus the interference of the variations of air mass and short-term local emissions can be eliminated and the influence of SA formation can be highlighted. The time window was fixed to two days for the convenience of intercomparing. We have added these descriptions into the manuscript as follow:

“These events were selected based on the similarity of $PM_{2.5}$ concentration and evolution, while the time window was fixed to two days for the convenience of intercomparing. In addition, during these events, the wind speed was generally low, the RH followed a general diurnal variations and SA mass grew steadily and continuously. Thus the interference of the variations of air mass and short-term local emissions can be eliminated and the influence of SA formation can be highlighted.”

4. L283-286, for me it is hard to tell from the data that the ratios were really lower after 4th Dec. Nor can I discern the “decreasing trends”.

Response: Thanks for your comments. It should be during the high RH events before 4th Dec when there were lower ratios and decreasing trends, and we have revised this sentence as “However, the ratios between N_{CCN} and mass concentration of $PM_{2.5}$ SA or NR- $PM_{2.5}$, were lower during the high RH period and demonstrated strong decreases, especially in Event 1 and 2.”

5. L294, by “the increase of hygroscopic particles”, do you mean number or mass concentration?

Response: It refers to the number concentration and we have revised it accordingly.

6. L304, is this statement necessarily true?

Response: Thanks for your comments. We have revised this sentence as “larger variation in CCN activity was observed in Events 3 and 4”

7. L311, by which metric do you define “CCN activity”? Do you refer to activation fraction?

Response: It refer to the size-resolved activation fraction rather than bulk activation fraction. The bulk activation fraction is determined by not only size-resolved activation fraction but also PNSD. Here we focus on particle hygroscopicity which is linked with particle chemical compositions and indicated by size-resolved activation fraction. We have revised the sentence as: *“Same as demonstrated in Fig. 2, SPAR was generally higher and thus particle CCN activity were generally stronger in high RH events than those in low RH events.”*

8. L313, *“the enhancement of particle CCN activity was stronger in low RH events”*, which metric or data is this statement based on?

Response: As mentioned in comment 7 above, “CCN activity” here refer to the SPAR as well. As shown in Fig. 3a, the difference between SPAR in high and low RH events at 300 nm decreased from 0.2 to 0.1 during the SA formations, indicating for a stronger enhancement in low RH events, probably due to both the stronger increase of SA mass fraction and the higher nighttime PA mass fraction (Fig. 2(e)). We have revised “CCN activity” to “SPAR” and added these description into the manuscript.

9. L319, it is not obvious to tell if there is *“the increases of Da”*.

Response: Thanks for your suggestions. The increase of Da is not significant and we have revised the sentence as *“This can be attributed to the strong increase of MAF and the slight increase of Da, which indicates significant increasing number fraction yet slightly weakening hygroscopicity of hygroscopic particles, respectively.”*

10. L321, again *“the enhancement of CCN activity was lighter”*, what metric or data is this statement based on?

Response: As mentioned in comment 7 above, “CCN activity” here refer to the SPAR as well. The enhancement of SPAR here refers to the description in Line 313 as shown in Fig. 3 (a). We have improved the description as *“Overall, the enhancement of SPAR was weaker but occurred at a broader particle size range in high RH events than in low RH events, as shown in Fig. 3a.”*

11. L325-327, *“unchanged CCN activity at low RH conditions”*, how is this statement drawn? Is this finding also valid for other SS?

Response: As mentioned in comment 7 above, “CCN activity” here refer to the SPAR as well. The discussion here is not necessary but may lead to confusion, thus have been removed.

12. L339, *“relatively smaller variations of particle density”*, this needs support from data or literature.

Response: Thanks for your suggestion. Based on measurements in the North China Plain, the variations of the accumulation mode particle density ranges from 1.2 to 1.8, whose relative variations are within 20% (Hu et al., 2012; Zhao et al., 2019). We have added this information into the manuscript.

13. L365, “*decreased continuously*”, it seems not to be a continuous decrease.

Response: Thanks for your comments. There is increase of NCCN(<300nm)/NR at early times of the SA formation before the decrease of NCCN(<300nm)/NR. So we have deleted “*continuously*”.

14. L445-447, it is not surprising that the correlation of NCCN_chem with NCCN_meas was not good as kappa was only derived from chemical composition of the bulk aerosol, which is highly biased to larger particles.

Response: Thanks for your comments. We agree that there may be significant deviations in the kappa estimated based on chemical composition of the bulk aerosol, which leads to significant deviations of NCCN prediction. However, in practice, the measurements of size-resolved particle chemical compositions are not common, and chemical composition of the bulk aerosol is still commonly applied in CCN studies (Zhang et al., 2014; Zhang et al., 2016; Che et al., 2017; Cai et al., 2018), especially when particle hygroscopicity measurements were in lack. In addition, we focus on the comparison between the different methods of applying the bulk aerosol chemical composition on NCCN calculation to provide a better method applicable for NCCN calculation on the NCP. We have added these descriptions into the fourth paragraph of section 3.2 as follow:

“Although there can be significant deviations for κ of accumulation mode particles derived from chemical composition of the bulk aerosol, which leads to significant deviations of NCCN prediction. However, in practice, the measurements of chemical compositions of accumulation mode particles are not common, and chemical composition of the bulk aerosol is still commonly applied in CCN studies (Zhang et al., 2014; Zhang et al., 2016; Che et al., 2017; Cai et al., 2018), especially when particle hygroscopicity measurements were in lack.”

15. L439-440, such a statement is not necessarily true. Primary particles can be CCN active. In addition, the authors defined kappa>0.1 as hygroscopic particle in the method part. Kappa of SOA can be <0.1, which contracts the statement here.

Response: Thanks for your comments. We agree that POA can be CCN active and kappa of SOA can also be lower than 0.1. However, in general, SOA have higher hygroscopicity than POA (Frosch et al., 2011; Lambe et al., 2011; Kuang et al., 2020). The statement here has been revised as “*As SOA is generally considered to be more hygroscopic than POA (Frosch et al., 2011; Lambe et al., 2011; Kuang et al., 2020c), the increase of hygroscopic particles or SA particles (both SIA and SOA) were*

considered to be the cause for the increase of SPAR within 200 to 300 nm size range (Fig. 2). In order to account for the variations of hygroscopic particles or SA particles in N_{CCN} calculation, Number Fraction of hygroscopic particles ($GF(90\%, 200\text{ nm}) > 1.22$, NF_{hygro}) measured by HTDMA and Mass Fraction ...”

16. L454, “real-time MAF can be estimated by MF_{SA} ”, how to estimate, by simple linear regression?

Response: The values of MF_{SA} were assumed to equal to MAF and used as real-time MAF to calculate SPAR and N_{CCN} . We have revised this sentence as “Thus, in the prediction of N_{CCN} , real-time SPAR can be calculated from average Da and MAF assumed to equal to real-time MF_{SA} (N_{CCN_MF}).”

17. L473, how do MAF and its diurnal variation depend on SS?

Response: The diurnal variations of MAF at the five measured SSs are shown as follow:

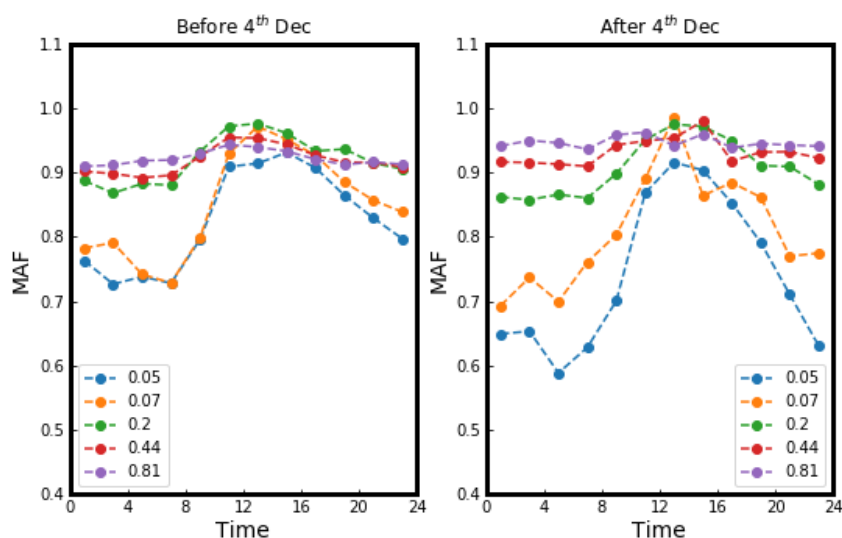


Fig. S5. Diurnal variations of MAF at the five measured SSs (indicated by different colors) during the high (left) and low (right) RH periods.

As mentioned earlier, the diurnal variations of MAF at the five measured SSs reveal significant diurnal variations in MAF at low SSs (0.05% and 0.07%) that are dependent on RH conditions, while weaker diurnal variations that are insensitive on RH conditions at SSs over 0.2%. In general, MAF become lower at lower SSs, especially during nighttime. We have added this figure into the supplements and this discussion into the last paragraph of section 3,2 (the summary of this section) as “The diurnal variations of MAF at the five measured SSs (Fig. S6) reveal significant diurnal variations in MAF at low SSs (0.05% and 0.07%) that are dependent on RH conditions, while small diurnal variations that are insensitive to the RH conditions at SSs over 0.2%. In general, MAF become lower at lower SSs, especially during nighttime.”.

18. L495, in the abstract, 50% was used while here 40% was used...

Response: Thanks for your comment and we have revised it accordingly.

19. L509, “mixing state”, is the right word here? What is the mixing state of these aerosols based on the “measurements of CCN activity, particle hygroscopicity and particle chemical compositions”?

Response: Thanks for your comments. Here “mixing state” refers to MAF (SPAR parameter). To avoid confusion, we have revised it to “MAF (SPAR parameter)” in this sentence and also in the abstract accordingly.

20. L797, in Fig. 2, it is helpful to describe the OA factors in the method part.

Response: Thanks for your suggestion and we have added the description as follow:

“including OA factors: hydrocarbon-like OA (HOA), cooking OA (COA), biomass burning OA (BBOA), coal combustion OA (CCOA), and oxygenated OA (OOA).”

Technical comments:

1. L214, “ NF_{hygro} ” was written as “ NF_hygro ” later.

Response: Thanks for your suggestion. We have revised them accordingly.

2. L272, “Dec.” should be “Dec”.

Response: Thanks for your suggestion. We have revised it accordingly.

3. L283, “are” should be “is”. “Higher” might be better than “stronger”.

Response: Thanks for your suggestion. We have revised them accordingly.

4. L324, “um” should be “ μm ”.

Response: Thanks for your suggestion. We have revised it accordingly.

5. L346, “normalized” is missed before $PM_{2.5}$? “Fig. 4(a1)” should be “Fig. 4(1a)”.

Response: Thanks for your suggestion. We have revised them accordingly.

6. L356, “of” should be omitted.

Response: Thanks for your suggestion. We have revised it accordingly.

7. L376-378, this sentence is hard to understand.

Response: Thanks for your comments. We have revised this sentence as “SA formation mainly

enhanced number fraction of CCN-active particles in particle size of 200 to 300 nm, as SPAR only revealed evident enhancement (Fig. S2(b2)) and N_{CCN} only significantly increased (Fig. 4(c2)) in that size range.”

8. L432-433, *“there were similar difference between CCN_AvgMAF and $NCCN_meas$ ” this sentence is hard to understand.*

Response: Thanks for your comments. We have revised this sentence as *“Only N_{CCN_AvgMAF} displayed similar deviations from N_{CCN_meas} , indicating that differences between N_{CCN_cal} and N_{CCN_meas} were mainly contributed by variations in MAF brought on by significant CCN-active particles number fraction growth due to SA formations.”*.

9. L452, *“the application of MF_SA on $NCCN$ calculation were shown”, it is in Fig. 6c rather than 6b.*

Response: Thanks for your suggestion. We have revised it accordingly.

10. L467-468, *this sentence is hard to understand.*

Response: Thanks for your comment. We have revised this sentence as *“Similar as before, NF_{hygro} was applied as a proxy for MAF in the N_{CCN} calculation, which also significantly improved the underestimation and correlation between N_{CCN_cal} and N_{CCN_meas} (Fig. 7(c)).”*

11. L825, *“the” should be “The”.*

Response: Thanks for your suggestion. We have revised it accordingly.

Reference:

- Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O. and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, *Atmos. Chem. Phys.*, 8(5), 1153–1179, 2008.
- Zhang, F., Li, Y., Li, Z., Sun, L., Li, R., Zhao, C., Wang, P., Sun, Y., Liu, X., Li, J., Li, P., Ren, G., and Fan, T.: Aerosol hygroscopicity and cloud condensation nuclei activity during the AC3Exp campaign: implications for cloud condensation nuclei parameterization, *Atmos. Chem. Phys.*, 14, 13423–13437, <https://doi.org/10.5194/acp-14-13423-2014>, 2014.
- Zhang, F., Li, Z., Li, Y., Sun, Y., Wang, Z., Li, P., Sun, L., Wang, P., Cribb, M., Zhao, C., Fan, T., Yang, X., and Wang, Q.: Impacts of organic aerosols and its oxidation level on CCN activity from measurement at a suburban site in China, *Atmos. Chem. Phys.*, 16, 5413–5425, <https://doi.org/10.5194/acp-16-5413-2016>, 2016.
- Che, H.C., Zhang, X.Y., Zhang, L. et al. Prediction of size-resolved number concentration of cloud condensation nuclei and long-term measurements of their activation characteristics. *Sci Rep* 7, 5819 (2017). <https://doi.org/10.1038/s41598-017-05998-3>
- Cai, M., Tan, H., Chan, C. K., Qin, Y., Xu, H., Li, F., Schurman, M. I., Liu, L., and Zhao, J.: The size-resolved cloud condensation nuclei (CCN) activity and its prediction based on aerosol hygroscopicity and composition in the Pearl Delta River (PRD) region during wintertime 2014, *Atmos. Chem. Phys.*, 18, 16419–16437, <https://doi.org/10.5194/acp-18-16419-2018>, 2018.
- Hu, M., Peng, J., Sun, K., Yue, D., Guo, S., Wiedensohler, A., and Wu, Z.: Estimation of size-resolved ambient particle density based on the measurement of aerosol number, mass, and chemical size distributions in the winter in Beijing, *Environ. Sci. Technol.*, 46, 9941–9947, <https://doi.org/10.1021/es204073t>, 2012.
- Zhao, G., Tan, T., Zhao, W., Guo, S., Tian, P., and Zhao, C.: A new parameterization scheme for the real part of the ambient urban aerosol refractive index, *Atmos. Chem. Phys.*, 19, 12875–12885, <https://doi.org/10.5194/acp-19-12875-2019>, 2019.
- Frosch M, Bilde M, DeCarlo PF, Jurányi Z, Tritscher T, Dommen J, et al. Relating cloud condensation nuclei activity and oxidation level of α -pinene secondary organic aerosols. *J Geophys Res- Atmos.* 2011;116(D22). <https://doi.org/10.1029/2011jd016401>.
- Lambe AT, Onasch TB, Massoli P, Croasdale DR, Wright JP, Ahern AT, et al. Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) activity of secondary organic aerosol (SOA) and oxidized primary organic aerosol (OPOA). *Atmos Chem Phys.* 2011;11(17):8913–28. <https://doi.org/10.5194/acp-11-8913-2011>.
- Kuang, Y., Xu, W., Tao, J. et al. A Review on Laboratory Studies and Field Measurements of

Atmospheric Organic Aerosol Hygroscopicity and Its Parameterization Based on Oxidation Levels.
Curr Pollution Rep 6, 410–424 (2020). <https://doi.org/10.1007/s40726-020-00164-2>