Dear Editor,

We greatly thank the reviewers for their review. Point-by-point responses addressing the reviewers' comments were uploaded (also attached to this file). The manuscript has been revised and improved accordingly.

Yours sincerely,

Jiangchuan Tao and Nan Ma

**Response to reviewers' comments on manuscript (acp-2020-939)**

(Reviewer comments in italics, the responses in plain font)

**Reviewer #1**

*The manuscript has improved a lot since the first version, therefore now I can imagine it being accepted for publication. However, I still have some comments and recommendations for the authors. And I still have to say, another language editing for the second half of the paper would be really nice. It is still very hard to read.*

Response: Thanks for your comments. Suggestions and comments are addressed point-by-point and corresponding responses are listed below. In addition, we have improved the language in the second half of the paper.

Specific Comments:

*Line 239: black carbon is assumed to be hydrophilic -> non-hydrophilic or non-hygroscopic*

Response: Thanks for your suggestion. We have replaced "hydrophilic" with "non-hygroscopic".

*Line 338-339: GF-PDF, SPAR is a function of the diameter, if you discuss an increase (or whatever) of these properties, please always indicate at which diameter you mean it*

Response: Thanks for your comments. Here we are referring to increase of SPAR in particle size range from 200nm to 400nm and increase of GF-PDF in GF range from 1.2 to 1.8. We have revised this sentence as: "*Simultaneous daytime increases in particle SPAR in particle size range from 200 nm to 400 nm, GF-PDF in GF range from 1.2 to 1.8 and SA mass fraction were found in all four events, suggesting that SA formation led to increasing hygroscopic particles number concentration, which in turn enhanced particle CCN activity.*"

*Line 343: was be found: please remove "be"*

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

*Line 361-386: Please discuss the fit parameters, the main parameters of the SPAR curves rather than the SPAR value at a certain diameter (at 300nm). Looking at Figure 3 (b and c) makes it clear what happens, and I do not read that from the paper, as it is written now. What I see there is, that Da values are almost exactly the same at low and high RH, which means that the hygroscopicity of this particle population is the same at high and low RH, during the SA formation there is for both cases a little* *decrease in Da which means a slight hygroscopicity increase due to the more hygroscopic SA material. There is a significant difference between the MAF values, but outside of the SA formation, so the fraction of the (as you call it) hygroscopic particles at low RH was lower in PA, and good part of this difference disappears during the presence of the SA formation. And with this the message for me would be, the hygroscopicity of the SA formed under low and high RH conditions is very similar, and the difference what you see in N_CCN is mainly because the mass/number and size of formed SA particles is different at low and at high RH.*

Response: Thanks for your suggestion. We have added the corresponding discussions in the end of this paragraph as follow:

"*In detail, the different variations of SPAR in high and low RH events indicated by MAF and $D_a$ shown in Figs. 3(b & c) suggested different variations of hygroscopicity, number fraction and size of SA particles. Before SA formation, there was a significant difference between the MAF in high and low RH events, which disappeared after the SA formation. The stronger variations in MAF in low RH events suggested stronger enhancement of number concentration of formed SA particles. As for $D_a$ during SA formation, there were similar, little decrease in both high and low RH events, suggesting similar hygroscopicity of the SA formed under low and high RH conditions. Thus differences of SPAR and the resultant $N_{CCN}$ during low and high RH events were mainly due to the different variations of number fraction of formed SA particles.*"

*Line 419-422: Please delete/correct these sentences, they are not correct. You cannot compare volume to number like this. You should compare N_CCN to the number concentration, that makes sense. But you have already discussed what happens with the SPAR value, which is exactly the same, the ratio of them.*

Response: Thanks for your comments. These sentences are incorrect and have been deleted.

*Line 431 and 342: within -> below*

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

*Line 441-451: Figure 3b shows that the hygroscopicity increase during SA formation was very similar in both low and high RH cases*

Response: Thanks for your comments. It should be the increase of number fraction of SA rather than SA hygroscopicity which leads to higher NCCN enhancement. We have revised the corresponding discussions in the manuscript as:

"*Under high RH conditions, there were strong SIA dominated SA formation leading to stronger enhancements of CCN-active particle number fraction and $N_{CCN}$. Meanwhile, under low RH conditions,* *there were moderate SOA dominated SA formation with moderate enhancements of CCN-active particle number fraction and* $N_{CCN}$.”

*Line 486: variation of MAF = changes in aerosol hygroscopicity is not true. You write that yourself one sentence before that the MAF represents the fraction of the hygroscopic particles. Please change this sentence.*

Response: Thanks for your suggestion. It should be the fraction of the hygroscopic particles and we have revised this sentence as “*Thus, for accurate* $N_{CCN}$ *estimations, considering the variation of MAF (changes in fraction of the hygroscopic particles) is highly essential.*”

*Line 487-494: Please let the reader know here, that this, what you talk about (trying to estimate the changes of the MAF from other measurements) here will be shown in the next part of the paper*

Response: Thanks for your suggestion. We have revised the last sentence in this paragraph by adding the corresponding description as: “*In order to account for the variations of hygroscopic particles or SA particles in* $N_{CCN}$ *calculation, in the following part, Number Fraction of hygroscopic particles (GF(90%, 200 nm)>1.22,* $NF_{hygro}$*) measured by HTDMA and Mass Fraction of SA particles* ($MF_{SA}$) *measured by ACSM in this campaign were used to represent MAF variations and to provide calculation of* $N_{CCN}$ *at SS of 0.05% …*”

*Line 495-497: please rephrase this sentence, very hard to understand*

Response: Thanks for your suggestion. We have revised it as: “*Based on the bulk hygroscopicity derived from particle chemical compositions measurements* ($\kappa_{chem}$)*, a critical diameter for CCN activation can be calculated based on κ-Köhler theory. With this critical diameter,* $N_{CCN}(0.05\%)$ *can be predicted incorporating measured PNSD* ($N_{CCN\_Chem}$).”

*Line 503: can be -> is*

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

*Figures 6 and 7: taking into account that MAF is less than 1 makes your prediction much better. What is not clearly stated, where is the Da coming from in this case? Please indicate it. It should be the chemistry/growth factor derived Da and not the one which was originally derived from the SPAR curves.*

Response: Thanks for your comments.

In figures 6 and 7, we evaluate the calculation of $N_{CCN}$ by using $NF_{hygro}$ or $MF_{SA}$ as the SPAR parameter MAF, which was found to contribute the most of the calculated $N_{CCN}$ deviation in Figure 5, especially during low RH periods. Besides neglecting the MAF variations, the differences between MAF and $NF_{hygro}$ (or $MF_{SA}$, shown in figure 6b or 7b) and the application of campaign average Da (Figure 5) can both contribute to the deviation of $N_{CCN}$ calculation. Thus, in order to highlight the application of using $NF_{hygro}$ or $MF_{SA}$ as estimation of MAF variations, the campaign average Da of SPAR curves in Figure 5 was used in the calculation of SPAR and $N_{CCN}$ in figures 6 and 7. As the deviation of $N_{CCN}$ calculation by using $NF_{hygro}$ or $MF_{SA}$ as MAF became smaller than those without considering MAF variations, it was concluded that $NF_{hygro}$ and $MF_{SA}$ can be used as MAF to improve $N_{CCN}$ prediction.

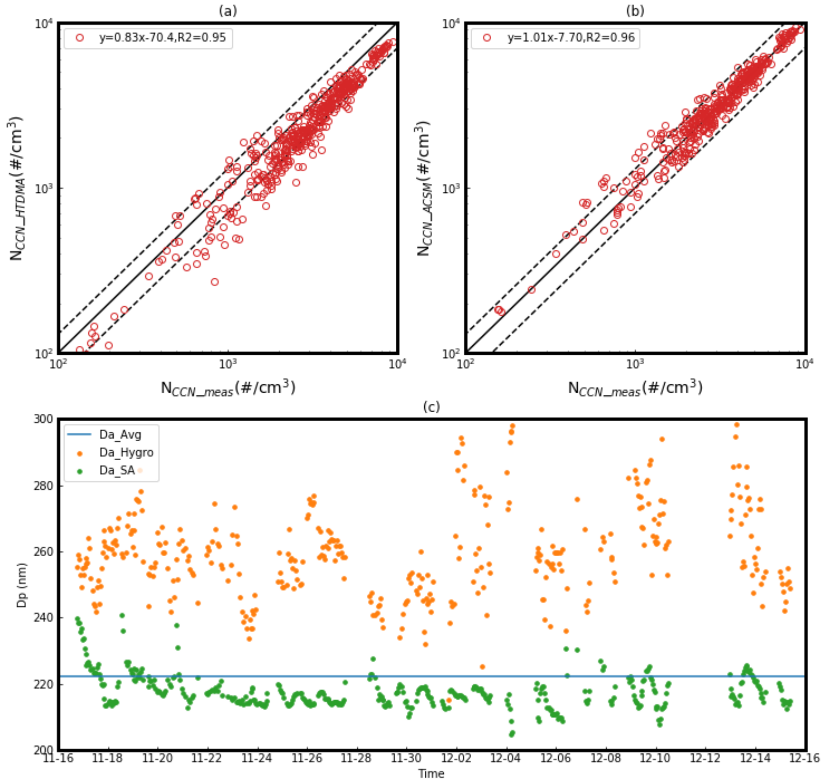

Figure R1. (a and b)The comparison between measured $N_{CCN}$ and calculated $N_{CCN}$ based on SPAR derived from real-time $MF_{SA}$ and GF derived $D_a$ ($N_{CCN\_HTDMA}$, a) or chemistry derived $D_a$ ($N_{CCN\_ACSM}$, b). The black dashed lines represent the relative deviation of 30%. (c) Time series of GF derived $D_a$ (blue markers), chemistry derived $D_a$ (yellow markers) and the campaign averaged $D_a$ from SPAR curves (green line).

In addition, we also agree that it's important to apply chemistry/GF derived Da in $N_{CCN}$ calculation as the reviewer suggested. However, as there can be significant deviations between CCN activity measured under different water vapor saturated conditions, there may be larger $N_{CCN}$ deviations by using real-time chemistry/growth factor derived Da rather than a campaign average Da, which can be obtained from other CCN measurement in the same areas. In the figure R1 shown above, $N_{CCN}$ calculated with chemistry/growth factor derived Da were compared with the measured $N_{CCN}$. As shown in figure R1(a), by using Da calculated from GF-PDF, the deviations of calculated $N_{CCN}$ ($N_{CCN\_HTDMA}$) become smaller, although there can be underestimation larger than 30%. This underestimation by using Da calculated from GF-PDF may be due to underestimating hygroscopicity of hygroscopic particles measured by HTDMA under sub-saturated conditions (higher values of yellow markers than the blue line in figure R1(c)). In Figure S3, similar underestimations (larger than 30%) of $N_{CCN}$ calculated with GF-PDF can also be found. In Figure R1 (b), the deviations of calculated $N_{CCN}$ chemistry derived Da ($N_{CCN\_ACSM}$) become smaller, although there can be overestimation larger than 30%. These smaller deviations may be due to better agreement between Da of SA (green markers in figure R1(c)) and campaign averaged $D_a$ from SPAR curves. However, this better agreement of Da may result from the combination of overestimated CCN hygroscopicity by neglecting CCN-active PA and underestimation of SOA hygroscopicity under super-saturated conditions. Furthermore, the difference of CCN hygroscopicity measured by different instruments which contributed to these $N_{CCN}$ deviations, was not the focus of this study. Thus, the chemistry/growth factor derived Da were not applied on $N_{CCN}$ calculation in this study.

We have made the descriptions of Da clear in the manuscript as follow:

Line 502-504, in the end of paragraph before figure 6 and 7*: "To be noted, in order to highlight the application of using $MF_{SA}$ as estimation of MAF variations on $N_{CCN}$ calculation, the campaign averaged $D_a$ from SPAR curves was used."*

Line 520-522, the paragraph for figure 6: "*Thus, in the prediction of $N_{CCN}$, real-time SPAR can be calculated from $D_a$ and MAF assumed to equal to real-time $MF_{SA}$ ($N_{CCN\_MF}$).*"

Line 538-539, the paragraph for figure 7:*"Also, the campaign averaged $D_a$ in Fig. 5a.was used to calculate SPAR curves and $N_{CCN}$.*"

Reviewer #2:

*The responses and the revised manuscript have largely addressed my previous comments. However, a few minor problems still remain.*

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

*1. Regarding my former general comment #1, I suggest the authors to explicitly specify supersaturation when describing NCCN in order to avoid ambiguity and over-generalization of the findings. For example, in the abstract (L36 and L570) and conclusion, NCCN can be changed to "NCCN(0.05%)".*

Response: Thanks for your suggestion. We have revised them accordingly and also made corresponding revisions throughout the manuscript.

*2. In the responses, Pg 34, the sentence of L4-6 were repeated.*

Response: Thanks for your comment. These sentences were incorrectly repeated in the responses, but the corresponding discussions in the manuscript have been confirmed to be correct.

*3. Regarding my previous specific comment #2, "L264, how are PA and SA characterized?" I meant in which method PA and SA were determined/quantified.*

Response: Thanks for your comment. PA (including four POA) and SA (including OOA and inorganic compounds) were determined based on ACSM measurement. In detail, OOA and four POA were analyzed based on the ACSM PMF analysis (Zhang et al. 2011). We have revised the sentence as "*The timeseries of meteorological parameters, SPAR,* $N_{CCN}$ *at SS of 0.05% and mass concentration of Non-refractory particulate matter of* $PM_{2.5}$ *(NR-*$PM_{2.5}$*),* $PM_{2.5}$ *SA (inorganic compounds and OOA) and* $PM_{2.5}$ *PA (primary aerosol, defined as the sum of POA) are shown in Fig. 1. The mass concentration of OOA and four POA were quantified by the ACSM PMF analysis (Zhang et al. 2011).*"

Reference:

Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Ulbrich, I. M., Ng, N. L., Worsnop, D. R., and Sun, Y.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, Analytical and Bioanalytical Chemistry, 401, 3045–3067, https://doi.org/10.1007/s00216-011-5355-y, 2011.