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Interactive comment on "Secondary aerosol formation alters CCN activity in the North China Plain" *by* Jiangchuan Tao et al.

Anonymous Referee #1

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The manuscript is about the influence of the secondary aerosol (SA) formation on the CCN activity based on a measurement campaign done on the North China Plane. The topic is very interesting, I would very much like to see a thorough study on it to get published. However, as the manuscript is now prepared, I have doubts about its quality, and in this form I cannot recommend it to be published in ACP. It needs a serious and thorough rework based on the referees' comments before it can be cosidered to be published. Please find my comments and remarks in the following.

General comments:

1. Too few events were analyzed in my opinion, to see whether really the change in RH cause a different CCN behavior. For such a study, more data would be needed than two short events for the high RH period and a single event for the low RH period. At least

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Discussion paper

use as many days for the data analysis as possible from this data set. For me, it looks like that you have chosen your RH criteria such, that only those days are included that you want to analyze even if there would be the possibility to include many more days when the RH was high or low. E.g. why don't you use 14th of December as a low RH event? Either use almost all the days with higher RH and lower RH for this comparison or do not do this low and high RH separation at all. Compare the campaign averages before and after the 4th of December, something like you show in Figure S2. As it is presented now, I am not convinced, that there is a significant difference between the low and high RH cases based on a solely 3 events. What if during the single low RH event something else than the RH caused the difference in the CCN activity? How can you be sure, that the RH is responsible?

2. Why do you only show the results at SS=0.05% when you have measured at 5 different SSs? Please show all the supersaturations you have measured. You could generally try to speculate a little bit less in the paper and at the same time show more important data, if you are afraid, that the paper will be too long. I know that you have mentioned, that you would like to focus on the low SS case, but you have still two other measured SSs smaller or equal then your upper limit of SS of interest (0.2%). Please at least include them in this paper. It would be nice to see whether SA formation have an effect on the CCN activity at those higher SSs as well or not.

3. At many parts of the paper, the MAF (maximum activated fraction) parameter appears (together with a single sigmoid fit) and is used for the fraction of the hygroscopic particles. As I mention later in the detailed comments, this parameterization/fit can only be used in certain cases. You should include a discussion and provide information on how well this fit could be used for your data. And dependent on the SS set in the CCNC, the MAF you present has a different meaning. You only show measurements at SS=0.05%, at this SS and with the highest considered dry diameter of 300nm, this MAF has the meaning of the fraction of the particles having a kappa at least approx. 0.22, far far away from non-hygroscopic. 1/3 AS and 2/3 BC would have such a kappa.

C2

Use MAF accordingly, and correctly! And I would need proof that this fitting method can be used for your data at any time during the campaign. For the 0.05% case, it assumes that there are no particle present around the kappa of 0.22, just a population with much higher kappa and a population with significantly lower kappa. Was it the case for the whole measurement period? If there will be other SSs included in the paper following my suggestion, then please check and show what the MAF would mean at that SS, like e.g. at 0.2% and maximum diameter of 300nm, the MAF would be the fraction of the particles having a kappa higher than approx. 0.013. Or a much better choice would be doing such a fit until a constant kappa at different SSs which would mean different maximal fit diameters. That would have a more useful meaning. Like the fraction of the particles having a hygroscopicity below kappa 0.1. That would mean that you have to use the measurements until a higher diameter than 300nm (approx. 390nm) at SS=0.05% which you did not include because of having too much noise. But that problem could be solved following another one of my previous suggestions and using more data and doing some time averaging. You have many choices, choose something which you like, but it is very confusing right now, and this MAF, as calculated now, is not representative for the fraction of the non-hygroscopic particles.

4. Something is strange for me for Figure 5a. How can it be, that the ratio between the calculated and measured N_CCN is systematically below 1? I would expect using the partly or completely averaged SPAR (whichever trace I look at it), that the ratio is scattered around one, but not being always below (like in Figure 5b). For me, this could only happen if e.g. you have a systematic error in the fitting procedure, which always underestimate the measured SPAR, or something else. In my opinion something can not be correct here. Please explain me, if the data is correct, how that can be.

Detailed comments:

Line72: "different with those" did you mean here different from those?

Line 86: hydrophobic is a too strong expression here, I guess you mean non-

hygroscopic

Line 132: how far was the container form the building of the gas measurements?

Line 139: you mention here the SS and the corrected SS of the CCNC, what is this correction? If it is simply the SS calibration, then you do not need to mention the wrong SS levels, just state the correct ones you determined based on the instrument calibration.

Line 154-156: about the inversion and multiple charge correction of the scanning CCNC system: you mention that a multiple charge correction was done and show some references, where details about it can be found. However, I really had to search longer among those papers until I found a method in one of them. Since the main result what you show in this paper is the SPAR, to my opinion the method of inversion/multiple charge correction has to appear a bit more detailed in this paper. And as I understood from the method I found in one of the references (if I found the method you used here), a simple correction only taking the multiple charged particles into account was applied. The width of the DMA transfer function was neglected. Please at least speculate on it, how much error you introduce to your measurement with this assumption.

Line 170 and 173: "under RH of 90%" please change under to at, under could be also understood as below

Line 179: you mention 4 dry sizes in Line 178 and then 6 sizes in this line. Which one is correct?

Line 209: What function was used for the fit?

Line 211: "(HGF?)" Typo?

Line 217: "(Da_hygro)" what is that?

Line 224: "dominate" change to dominant

Line 240: please change "reported in the same…" to "reported from the same…"

Line 243: what are the kappa values you used for the inorganics? The kappa theory is not a perfect parameterization of the water activity, and therefore it is not granted that a kappa you calculate from a HTDMA will be the same as what you get from a CCN measurement. For example, AS has a different kappa at supersaturation and at 90% RH. How did you take this into account? Please comment on it. And at what RH was the relationship for kappa_org determined in the mentioned study? Line 248: it is not generally parameterized, often but for sure not generally, please correct Line 249: change hydrophobic to non-hygroscopic, or what kind of hydrophobic particles do you mean? I am not aware of any kind of atmospheric aerosols that are hydrophobic. To my knowledge non-hygroscopic (kappa=0) aerosol particles activate like a completely non-soluble but wettable surface according to the Kelvin-effect. Hydrophobic particles activate at even worse than those, so at a higher SS.

Next to that, a CCNC can theoretically measure non-hygroscopic activation at any SS, you simply need to get to a high enough particle diameter. So please change the sentence accordingly mentioning, that your used setup, which only goes up to 300nm, was not able to capture the activation. Next to it, at your highest SS of 0.8%, non-hygroscopic particles (kappa=0) that have larger dry diameter than 270nm already activate. So, at your highest SS and diameter of 300 nm you should activate the non-hygroscopic particles as well and get an MAF of 1 (assuming now a very narrow DMA transfer function which might not be the case) independent on the fraction of the non-hygroscopic particles. Line 254: "can represents" do you mean here can represent or represents? The later would not be true, if you have a hygroscopic fraction of the aerosols with not a single kappa but a broader kappa distribution. Please include a discussion on this here.

Line 255: sigma of the error function: does not only include the heterogeneity of the hygroscopicity but also the transfer function of your measurement system, mainly the DMA transfer function.

Line 257: see my previous comments on "hydrophobic". Kappa<0.1 is not even non-

hygroscopic. It would be something like a particle consisting of approx. 17% of AS and 83% of BC. One definitely cannot call this hydrophobic.

Line 266: change "is" to "was"

Line 290: you show the CCN activation ratio/fraction not activity Section 3.2: From figure 1 it looks like, that you have a strong diurnal variation of the CCN activity almost every day. Somehow you only show the results of the few selected events. Please show at least an average (and the variation) of all the days for the data you show in Figure 2. And discuss them. It would be also nice to show the diurnal variation of the number size distribution as well.

Line 301: please correct "hydrophobic"

Figure 3a: please include the standard deviation of the averages for the SPAR curves as error bars or shading

Figure 3b-c: what are the error bars? The error of the fits, or the standard deviations of the calculated averages, or something else?

Line 327-329: I do not understand this sentence

Line 331-33: I do not understand either

Line 348: "to can be expected" typo

Figure 4b: showing the number of aerosol particles instead of the volume would be much useful, the CCN activity is also measured by the number and not by the volume

Line 399-402: Sentence too long, please start a new sentence after "respectively" and reformulate if, it is hardly understandable.

Line 411: change please "was" to "is"

Line 420: do you mean "is calculated based on. . ."?

Line 420: CCN activity is not a quantity, somehow you use that through the whole paper

as it was. Please correct it everywhere. What do you mean by it here? The SPAR? Or some kind of N_CCN? How is the N_CCN_cal exatly defined? Or is that the calculated N_CCN? Please rewrite this whole sentence and explain how you exactly calculated the CCN prediction.

Line 426: "as to" -> "to as"

Line 439-442: For me it would be strange if using a completely different instrument for a kappa measurement from bulk chemistry assuming internally mixed aerosols would improve the N_CCN prediction compared to the prediction based on the averaged SPAR. Please do not introduce this prediction method as an improvement.

Line 440: please include the exact definition of the number fraction of hygroscopic particles!

Line 453: calling R^2=0.59 a "strong correlation" is maybe a little bit too strong.

Figure 6: Please show the calculated vs. measured N_CCN for the methods you used for Figure 5 as well to have a comparison.

Line 459-472: you could not only use the bulk HTDMA hygroscopicity but the complete GF-PDF for the N_CCN estimation considering the mixing state of the aerosols as well. For sure, that would improve the calculation as well.

Line 473-478: If you want to show the importance of the changing MAF in the N_CCN prediction then you do not need all these calculations using the HTDMA and the AMS and the MAF prediction based on a whatever measured parameter of these instruments. Just simply show the calculated N_CCN (averaged MAF) vs the measured N_CCN (MAF as it was measued) as you calculated for the orange line in Figure 5. And as it looks like from Figure 5 you would not have an average error higher than 10% using the averaged MAF, so I am really not convinced about your summary statement. It might be important to take an average MAF different from 1 into account, but most probably not its time variation.