

## Review report on Wang et al. 2022, ACPD

The authors present a very thorough description of a 4 year-long size resolved CCN measurement data set at a central European rural background station. Due to the long dataset, the authors can analyse seasonal trends and test multiple approaches to predict the number of activated particles  $N_{CCN}$ . This type of data and analysis approach is very valuable for the atmospheric science community. But unfortunately, the authors mostly simply present and describe the measurements and leave the reader wondering what causes these trends and what they may mean in the big picture (e.g., for climate modelling). Adding these necessary discussions constitutes major revisions. I recommend publication after these are done, and the remaining issues listed below have been addressed.

### Main Comments

- 1) Throughout the manuscript, the balance between describing the measured values and interpreting them is strongly tilted towards the descriptive side. This data set contains a lot of interesting scientific observations. But in most places the reader is left to wonder what they mean. And if interpretation is provided, it is hidden after all those detailed numbers which are already visualised in the Figures/Tables. By describing every detail, the authors lose the attention of the reader for the really interesting parts. Below is one example to illustrate this issue.

In Chapter 3.2, the authors describe and name literally all values of  $k$ ,  $D_c$ , and  $(D_{75}-D_{25})/D_c$  which are written down in Table 4 and shown in Fig 6 (lines 342-376 = 34 lines). Then they provide 8 lines stating that aerosol is least internally mixed in winter with 1 very short sentence saying that that is potentially from local pollution. This is then followed by a general explanation how their type of CCN measurements probe different parts of the particle size distribution (7 lines). This information should have been provided at the start of this chapter as it applies to all values presented here. Next are two paragraphs (9 and 10 lines each) briefly describing the results presented in Fig 7 and Table 5, fortunately in a general way and without providing each individual number.

After these lengthy descriptions, I ended up with the following questions:

- a) why is  $k_{CCN}$  smaller than  $k_{chem}$ ?
- b) why is the aerosol more internally mixed in summer?
- c) what is that local pollution that is a plausible explanation and why is it only relevant in winter?
- d) why is  $k_{CCN}$  more sensitive to  $D_p$  in winter?
- e) my answer to (c) is: because the winter aerosol is less internally mixed, especially for small particles. Then I wonder: Why are the small particles less externally mixed?
- f) and following that: why are the trends in  $k \sim D_p$  and  $(D_{75}-D_{25})/D_c \sim D_p$  different? Both describe the degree of externally mixing.

None of these aspect are picked up anywhere. Yes, the reader can come up with their own answers but that is not the point of such a manuscript.

- 2) The manuscript feels massive in its current state. In addition to the lengthy descriptions (which need to be shortened), there are 11 Figures and 5 Tables. The author should consider which of these are really needed to follow the main story line in their manuscript. Currently, the amount of information is a bit distracting from the main points. In my opinion, the following things can go to the Supplement Information without compromising the content of the manuscript: Fig 1, Fig 2, Fig3a, Fig 5, either Fig 8 or Fig 9, Fig 11, Fig A1, and Table 3 and 5. Table 2 can go to SI as well if the authors follow my suggestion in Specific Comment 15. This together with trimming the detailed descriptions, will make this paper a lot more reader friendly and highlight the interesting scientific findings while still providing all the details in the SI.
- 3) The authors investigate the mixing state of the aerosol particles. While the definition of the term “internally mixed” is clear (all particles at any given size have the same composition, Seinfeld&Pandis), there are two aspects of external mixtures. It can mean that at a given size, particles can have different compositions. This shows up in size-resolved CCN measurements as a broadening of the AF vs  $D_p$  function ( $(D_{75}-D_{25})/D_C = IQR/D_C$ , IQR: interquartile range). On the other hand, externally mixed can also mean that the composition varies at different sizes (e.g., large dust particles and small SOA particles from NPF). If the CCN measurements probe different parts of the size distribution, the  $k_{ccn}$  values will be different according to the size dependent particle composition. The width of the AF vs  $D_p$  function can also be affected in this case. In their investigation, the authors only use the  $IQR/D_C$  values to infer information about the mixing state and did not link this information with the apparent size dependence of  $k_{CCN}$  and the discrepancy to  $k_{chem}$ . The manuscript will benefit from adding some discussion of these two manifestations of external mixing (see also Specific Comment 29)
- 4) The authors claim that their findings (i.e., their parameterisation of  $k \sim D_p$ ) can be applied to other locations with a “similar aerosol background”. But they do not qualify what they mean by that. How would one determine the similarity? From CCN measurements? Or can composition measurements be used? Fig 10 suggests that Melpitz is more similar to Shanghai than Barbados or the Amazon. So, the categories rural/urban may not always be helpful to identify “similar” behaviour. Discussing what needs to be similar to enable the application of the  $k \sim D_p$  parameterisation should be a section in the conclusions chapter.

## Specific comments

- 1) line 56 ff: This sounds like Petters and Kreidenweis (2007) came up with a new formula instead of the Köhler theory. But they just parameterized the Raoult term with a single parameter to capture the water activity without needing to know anything about the dissolved compounds. Adjust the sentence to reflect that.
- 2) line 64: "...because of the reciprocal relationship between  $k$  and  $D_p^3$ " The word reciprocal is ambiguous in this context. But mainly, I disagree with this statement. The stronger impact comes from the different range in which  $k$  and  $D_p$  vary and that  $D_p$  is cubic. The maximum variation for  $k$  is  $\sim 0.05 - 0.8$ . That is a change of factor 16. But the dominant particle size may change from 30 nm to 200 nm.  $200^3/30^3 = 296$ .
- 3) line 156: 40% RH seems still quite high for size selected CCN measurements. For aerosols with a high hygroscopicity, there may be considerable amounts of water in the selected particles which would then create a noticeable bias in the measured  $D_c/SS_c$  pairs.
- 4) line 162: What is meant by "near-PM1"?
- 5) line 177: The total SS cycle is 2.5h. Were all SS steps of the same length? Was all data used or were the first x min omitted while SS stabilised after change? Was there one  $D_p$  scan or multiple during one SS setting?
- 6) line 184: Rose et al. (2008) point out that you need to state which parameterisation/model was used to derive the theoretical SS/Dcrit values. They provide many in their study. Which one was used here? Also, what does "regularly" mean? Every month?
- 7) line 207ff: What was the size range of the DMA coupled to the CCN-C? Later in Line 391, the authors state that the limit was 40-200nm for  $D_p$ . This information needs to go into the Methods section. Was the D-MPSS compared with the DMA-CPC to check that the particle losses in each instrument are comparable? Also, why was the upper size limited in the DMA? Was it due to HV? Or was there a physical limit (Impactor, too many bends in line?). I.e., are you sure that the large particles did make it through the DMA in the same way as they reached the D-MPSS?
- 8) Line 210f/ Eq 1: What is meant with "a and b are the lower and upper limits for calculating  $D_c$ "? To my understanding, 'b' is the height of the upper plateau of the sigmoidal function and 'a' is the offset from 0 in the y direction. If b is lower than 1 (after accounting for any instrument discrepancies), this indicates the presence of particles that do not activate (e.g., pure black carbon or dust). As the mixing state is investigated later on, it would be good to know if the b value was lower than 1 during the times with higher external mixing.
- 9) Line 227 ff: As stated in Petters and Kreidenweis (2007), Eq 2 is an approximation which is only valid if the solution is very dilute at the point of activation. Is this approximation true for the analysed data in this study? I.e. are the kappa values high enough (Petters and Kreidenweis (2007) suggest  $k > 0.2$  as the threshold)?

- 10) Line 230: The authors assume that the surface tension of the droplet is constant, and  $k$  is the only unconstrained parameter in their equation. If the surface tension does vary, the effect in measured aerosol particle activation behaviour will then be accounted for by a different  $k$  value. Then  $k$  is not just representing the hygroscopicity of the aerosol but also the effect of any surface tension change and may differ more from  $k_{\text{chem}}$  which can only take the composition into account. Since the debate about the importance of surface tension changes and the connected bulk/surface partitioning is still ongoing (e.g.: Ovadnevaite et al. (2017); Vepsäläinen et al. (2021)), I recommend that the authors simply include a brief sentence stating that  $k$  may pick up surface tension changes.
- 11) Line 242: Why was  $k=0.1$  used for organics? In your introduction, you show that  $k_{\text{org}}$  can vary enough to be relevant for  $N_{\text{CCN}}$  predictions (e.g., in the range of 0.05 – 0.15 which can be caused by changes in organic composition). Ambient and chamber measurements have shown that  $k_{\text{org}}$  is a function of the degree of oxidation (e.g., expressed with O:C). What impact will it have if you use such a parameterisation for  $k_{\text{org}}$  (e.g.,  $k_{\text{org}} = 0.18 \cdot \text{O:C} + 0.03$  given in (Lambe et al., 2011))
- 12) Line 245 “When  $k$  is given...” For equation 4 to work, only a value for  $D_{\text{C}}$  is needed. Size resolved CCN counter measurements provide SS/ $D_{\text{C}}$  pairs and Eq 4 can be used directly. If no  $D_{\text{C}}$  values are available,  $D_{\text{C}}$  can be calculated for any SS from a given  $k$ .
- 13) Average values in Fig 3, 4, 5, and 7: The authors show average values of  $N_{\text{CCN}}$ , AR, etc. and use the standard deviation to indicate the spread of the data. Would it not be better to use the interquartile range for that purpose? So, indicating the mean (or median) value with a marker and then use a error bar/shaded area to indicate the Q25 and Q75 range?
- 14) There are multiple issues with Fig 3.
  - a. The Figure caption does not contain the information about the black markers. in Fig 3a. There is no description of the right-hand axis in Fig 3a.
  - b. Neither is there any information about the averaging which seems to be different for the size distribution (two values per month?) and the AR values.
  - c. The panels in Fig 3a are so tiny that it is very hard to see, e.g., the differences between SS=0.5% and 0.7%.
  - d. The error bars are outside of the y-axis range for some plots in Fig 3b and c.
  - e. For me, Fig 3a added very little to understand the description provided in Chapter 3.1. Also, for the overall interpretation of the data, the CCN number size distribution is not as relevant and could easily move to the SI. See next comment for other changes proposed for Fig 3.

15) The description of the AR and  $N_{CCN}$  values and trends is very hard to follow in Chapter 3.1. This is caused by the excessive details in the description and the choice of visualisation of the data.

- a. It is good that the authors compare their values to so many other studies. But due to each study having a different SS range, it is difficult to really understand how the data compares. Table 2 does not provide much more insights. But a simple Figure does (Fig R1 below):

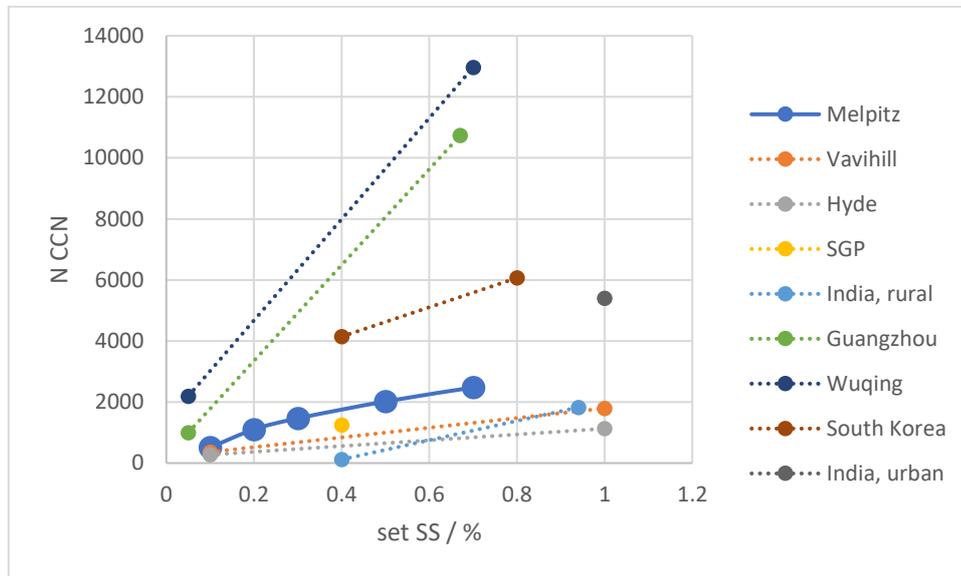


Figure R1:  $N_{CCN}$  from this study and studies cited in chapter 3.1.

- b. The authors look into the seasonal trends of  $N_{CCN}$  and AR and again I got lost in all the numbers and the provided visualisations do not help. Looking at the plots in Fig 3b and c, the seasonal behaviour of AR and  $N_{CCN}$  look indeed “similar”. But when I visualised the given values from lines 273 – 280 as a function of season (see Fig R2 below), I realised that there are some interesting differences. Going from 0.1% to 0.7%, the minimum of the  $N_{CCN}$  shifts from autumn to summer (Fig R2 top) while the trends in AR with a minimum in summer are the same for these SS (Fig R 2 bottom. Why is that the case? To me, it is clear that this must be connected to changes in the PNSD. But before the authors get to that, they first dive into the details of the  $N_{CCN}(SS)$  and  $AR(SS)$  relationships. Here already the connection between the set SS and the size range that is probed is important. But that is not mentioned until lines 385-391 (see Specific Comment 28)

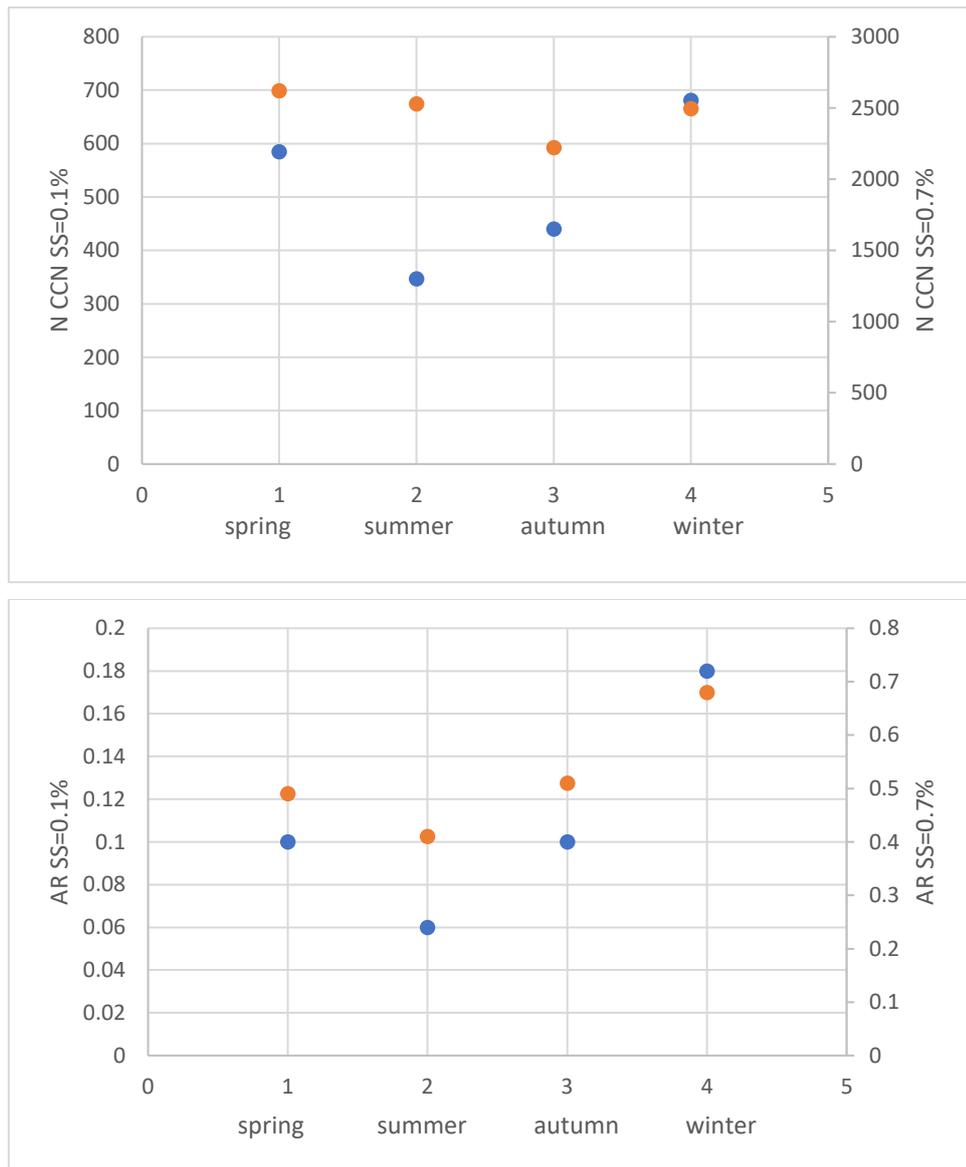


Figure R2: Seasonal trends of  $N_{CCN}$  (Top) and AR (bottom). Values for SS=0.1% are in blue and use the left-hand y-axis, while the values for SS=0.7% are indicated in orange and use the right-hand axis.

- c. I do not understand what the authors mean with “CCN number size distribution gradually peaks in summer”. Whatever is meant by that, how is that connected to the seasonal trends in  $N_{CCN}$  and AR, especially the summer minimums of AR and  $N_{CCN}$ (SS=0.1%)?
- d. Why are AR and  $N_{CCN}$  more sensitive to SS in summer than in winter?

My recommendation for cleaning up this chapter is to change Fig 3 by moving Fig 3a into the SI. Instead, provide a larger version of the “all data set“  $N_{CCN}$  vs SS which includes the values for the other studies as shown here in Fig R1. This will make the naming of all the number for the previous studies obsolete and Table 2 can also be moved to SI. If the authors keep the description and deepen the discussion of the seasonal trends, I strongly recommend adding something like Fig R2 either to Fig 3 or into the SI to help the reader follow the descriptions. Shifting the explanations

about the PNSD to follow the description of these trends will then feel more natural. The next few Specific Comment are also related to improving this chapter.

- 16) line 282ff: The error function used for fitting seems to have 4 free parameters. Each data set that is fitted has 5 values. Some people might say that that is a problem. Or was the original data fitted and not the (seasonal) averages? This is not mentioned in the text.

However, since the error function fits are not used anywhere in the manuscript other than stating that the function fits slightly better, this could be reduced to stating that the fit was also performed with an error function and the fitted parameters and curves are in the SI.

- 17) line 298: This information about what the CCN number size distribution represents should have appeared at the end of the methods section when EQ 4 is introduced. The schematic diagram is using the assumption that there are no non-CCN active particles at  $D_p > D_c$  (see also Specific Comment 8). Is that assumption reasonable? You should be able to estimate that from checking the plateau values in the AF vs  $D_p$  plots. How close are these values to 1.0 (after you accounted for different losses in the two instruments/sampling lines)?

- 18) line 314f: Again, the phrase “CCN number size distribution gradually peaks in summer” occurs without clarifying what is peaking.

- 19) Lines 306-317: It seems plausible that the presence of a large number of small particles explains the minimum of AR and maximum of  $N_{CCN}(SS \leq 0.5\%)$  during summer. But why was the influence of the change in hygroscopicity omitted? Winter and Spring have much higher  $k_{CCN}$  values (at least for the larger particles) which will also contribute to the high AR and  $N_{CCN}$  during that season.

- 20) Fig 4 should be improved. The black markers for  $k_{chem}$  are difficult to see in front of the dark blue background from  $NO_3$ . Why does the  $k_{chem}$  axis start at 0.2 and not 0? I do not like how the GMD and  $N_{aero}$  plot are put over the PNSD graph. Overlaying two panels over the PNSD is not straight forward to read. The intuitive interpretation is that the two sets of black markers are both on an axis extending the full height of the PNSD plot. The March and February markers are only partially visible.

- 21) Lines 323-329: While these correlations are interesting, Fig 5 could also be in the SI. Especially, since there is no interpretation of the meaning of the  $k \sim fX$  correlations currently. Yet another example where the reader is left to come up with their own conclusions about an interesting observation. Here is my take:

For understanding the relationship between  $k_{chem}$  and the individual composition groups, it is important to realise that these groups do not act in the same way in Eq 3. The influence of Org is direct.  $k_{org}$  is smaller than  $k_{inorg}$ . Thus, higher  $f_{org}$  means lower  $k_{chem}$ . But with  $SO_4$ ,  $NH_4$ ,  $NO_3$  it is more complicated because they are coupled through the ion balance. The absolute amount of  $SO_4$  and  $NH_4$  seems pretty stable. But the  $NO_3$  amount changes a lot between the seasons. The presence of  $NO_3$  shifts the salts from mostly  $(NH_4)_2SO_4$  towards  $NH_4NO_3$  and  $NH_4HSO_4$  or even  $H_2SO_4$ .  $k$  values are very similar between  $(NH_4)_2SO_4$ ,  $NH_4HSO_4$ , and  $NH_4NO_3$ , but  $k_{H_2SO_4}$  is much higher. Thus, an increase in  $NO_3$

can have a dual impact on  $k$  for this data set. The increase in  $\text{NO}_3$  adds a higher proportion of salt and also increases the  $k$  of  $\text{SO}_4$ .

So, if  $f\text{SO}_4$  decreases because more Org is present,  $k$  decreases. If  $f\text{SO}_4$  decreases because more  $\text{NO}_3$  is present,  $k$  increase. As these two trends are opposite, the correlation of  $f\text{SO}_4$  and  $k$  will be poor. Since this behaviour is opposite the usually assumed “ $f\text{SO}_4$  increase leads to  $k$  increase”, it is worth discussing. Also, why is  $\text{NO}_3$  increasing in winter and spring? Can it really be just the change in ambient  $T$ ? Could it be linked to the “local pollution” that is mentioned without any explanation in line 384? And to link this to the bigger picture: could the balance between  $\text{NO}_3$  and the other aerosol constituents be an important factor when comparing aerosol activation behaviour in different regions?

- 22) Lines 330-337: The changes in the width of the CCN number size distribution are not just related to the hygroscopicity (i.e., the  $D_c$  values). The shape of the PNSD plays an equally important role. The  $D_c$  value (i.e. the hygroscopicity) determines the edge at the smaller end of the CCN number size distribution. But the shape of the distribution at larger sizes depends more on the shape of the PNSD. I.e. with an identical  $k / D_c$  the winter CCN number size distribution will be wider because of the shift to larger sizes in the PNSD. The different shape of the PNSD may also help to explain the stronger sensitivity of  $N_{\text{CCN}}$  to SS during summer. The PNSD is probably steeper in the 40-150nm size range. Thus, a small shift in  $D_c$  will change the  $N_{\text{CCN}}$  much more than in winter where the PNSD look broader.
- 23) Chapter 3.2: Throughout the manuscript the authors write as if  $k_{\text{CCN}}$  and  $D_c$  are independently measured variables, while really  $k$  is calculated from the measured  $\text{SS}_c/D_c$  pairs. Describing both the  $D_c$  and  $k$  trends in details is thus redundant. As the authors want to compare the hygroscopicity to the composition, it is sufficient to present the  $D_c$  values only in the Table and figure. If a reader is interested in the exact values for  $k_{\text{CCN}}$  or  $D_c$ , a Table/Figure is anyway much faster than trying to find the relevant values in the long text. Then focus on the  $k_{\text{CCN}}$  trends and compare them with the  $k_{\text{chem}}$  information (see also next Specific Comment). This will make the actual discussion/interpretation/comparison much more readable. To facilitate the  $k_{\text{CCN}} / k_{\text{chem}}$  comparison, add the  $k_{\text{chem}}$  values to Fig 6a.
- 24) Lines 347f: I disagree with the statement that the seasonal variation of  $k_{\text{chem}}$  and  $k_{\text{ccn}}$  are similar for all SS. Adding the  $k_{\text{chem}}$  values to Fig 6a would make this clearer. The season trend in  $k_{\text{ccn}}$  is much weaker for small particles and I would claim that  $k_{\text{CCN}}(\text{SS}=0.7\%)$  does not display any trend if its values are 0.19, 0.20, or 0.21, each with a standard deviation of 0.1. This is already a strong sign of a more externally mixed aerosol population during winter and spring. It also shows that  $k_{\text{chem}}$  is not representative for the smaller particles at this location. See also Specific Comment 28 and 29.
- 25) Line 339f:  $k$  also varies with composition!
- 26) Line 379: “non-urban locations” Is the point here that the particles are away from strong localised sources? Or is this about the type of aerosol (e.g., anthropogenic vs biogenic)?
- 27) line 384: This is another example where the manuscript has a lot of description and very little discussion. What is that local pollution? How does it explain the observations? Would this local



pollution have varying effects depending on the particle size? Why is this pollution more important in Winter?

- 28) lines 385-391: This important explanation needs to come much earlier in the text since it is not only relevant for the  $IQR/D_c$  vs  $D_p$  relationship but also for all discussion related to the size resolved CCN measurements. Especially, when comparing with  $k_{chem}$ . As the ACSM is sensitive to mass (and not number) concentration, the bulk composition is dominated by the contribution of the larger particles. Thus,  $k_{chem}$  may not be representative for the smaller particles (higher SS) which is exactly what Fig 6 shows.
- 29) Lines 392-410: Here the authors again just describe the observations without making the interesting connections. The authors do not draw the connection between the change in  $D_p$  dependence of  $k_{CCN}$  and the change in mixing state (see also Main Comment 3). The higher sensitivity of  $k_{CCN}$  to  $D_p$  during spring and winter is not an intrinsic property, but it is the direct result of a more externally mixed aerosol population with size dependent composition. In spring and winter, it is more important which part of the size distribution is probed by the CCN measurement because the particle composition changes more with size than in the other seasons. Now, the authors should think about why this is the case? What causes this stronger size dependence of the particle composition? And what does it mean that the  $IQR/D_c$  vs  $D_p$  relationship is much shallower in spring than in winter?
- 30) Chapter 3.3: The introduction of the prediction methods is currently a little bit confusing and needs improvement. From the text, I did not understand what the main difference is between the two categories. I eventually work out that the schemes in the first category can be used for data obtained from polydisperse CCN measurements when only  $N_{CCN}$  is measured while the second category is based on using some sort of  $k$  value to calculate  $D_c$ . In addition, readability could be enhanced by labelling the schemes using the categories, e.g., N1, N2, and K1, K2, K3.
- 31) lines 439f: RD is a single value for each case in Fig 8. But  $|predicted N_{CCN} - measured N_{CCN}| / measured N_{CCN}$  provides a number for each measurement point. I guess these values were summed? Please, correct this equation and write it as its own as a proper equation and not “in-line”
- 32) Lines 443-454 summarise the prediction quality of the 5 schemes. This section is good. But then that is followed by yet another very detailed description of numbers that are presented in Fig 8 and 9 (over 3 and a half pages!). This is extremely tedious to read and again the important conclusions are buried under mountains of numbers. The authors need to trim this section.

Fig 8 and 9 show the same information simply from a different angle. They need to decide which of the figures works better and put the other in the SI. Instead of providing so many numbers for each scheme to say again that the prediction is better/worse, they should focus on the main improvements and features of the schemes which lead to the better/worse prediction. E.g., scheme 1 calculates 1  $N_{CCN}$  value for each SS. Thus, the spread depicted by the boxplot in Fig 9a simply reflects the standard deviation of the measurements as shown in Fig 3c (or rather the Interquartile range). For the category 2 schemes, the point is how well the parameterised  $k$  describes the measured  $k_{CCN}$  value. This then

explains why some seasons are predicted better than others (i.e. if the measured  $k_{CCN}$  are closer to the value set in the scheme).

- 33) lines 510 – 528 provides a good summary of the performance of the schemes and links the power law values to other observations. But what does it mean that the values for Melpitz are similar to some stations (even urban ones) and not to others (see also Main Comment 4). Either here – or better in the conclusions – this should be discussed, and the authors should at least speculate what may be causing the similar behaviour at such different sites.
- 34) Table 3 is very difficult to read. It is next to impossible to compare the parameters as each entry is spread over multiple lines. How about stating the equations in the Table caption and then providing only the parameters and  $R^2$  values in the table. If this table stays in the main text, the error function values should be moved to the SI (see Specific Comment 16).
- 35) The authors claim that scheme 5 (using the power law  $k(D_p)$  approximation) provides an improved prediction of  $N_{CCN}$ . This is true when compared to schemes 1 and 2. But how much does that improvement really matter when looking at schemes 3-5? From 4 to 5, the slope decreases 0.1 on average. So, the 10% overestimation is reduced. How much will that impact, e.g., the calculation of radiative forcing or prediction of precipitation in a climate model? Is that worth the effort? Some people may argue that operating an ACSM from which  $k_{chem}$  can be derived, is more feasible in a measurement station than conducting size resolved CCN measurements which are needed to obtain the  $k(D_p)$  relationship.
- 36) The  $k$  value used in scheme 3 is clearly too high. Have the authors tried to run this scheme using the average  $k_{CCN}$  value for their data set? How “good” is scheme 3 then?
- 37) The Conclusions chapter is simply a summary of the previous chapters, repeating many of the numbers that were already stated. These are not “conclusions” as in interpretations or putting their findings into context. There are many things the authors bring up in this chapter. These are a few ideas that spring to my mind (some are already mentioned in other Specific Comments):
  - a. How much their improved  $N_{CCN}$  prediction may improve modelling results?
  - b. How much would using the values from the “wrong” season affect  $N_{CCN}$  predictions? or from a wrong location (E.g. using the Budapest values for the Melpitz data set)
  - c. If the  $k \sim D_p$  prediction works so well, do we really need continuous CCN measurements? Wouldn't it be enough to determine the representative  $k \sim D_p$  fit for a few representative locations?
  - d. Or playing devil's advocate: Since the  $k_{chem}$  based  $N_{CCN}$  prediction is much better than the ones based on  $N_{CCN} \sim SS$  or  $AR \sim SS$ , wouldn't it be better to improve composition measurements?
  - e. regarding the mixing state: Why is the mixing state different between seasons?
  - f. Why is  $k(DP)$  and  $IQR/D_C(D_p)$  different between the seasons?
- 38) line 565: these things are also linked to the highest  $k_{CCN}$  and the widest spread in  $k_{CCN}$  (i.e., least internally mixed)

## Language:

General: In multiple locations, main clauses are attached with “;” to each other. While this is grammatically possible, it decreases readability by creating “monster sentences”. Simply use a full stop and start the second main clause. Examples: line 28ff: second sentence starts at “the seasonal mean activation ratio...”

line 15 “measurements on aerosol particle activation” -> of

line 20 “improving predictions”: predictions of what?

line 29 “twice higher” -> either “twice as high as” or “two times higher than”

line 35: “the power law function” sounds as if this is a specific function with the name ‘power law’ change to “a power law function”

line 44 “activated cloud droplets” -> remove activated. The particles get activated to grow to cloud droplets.

line 72f “should be underlined” -> no, it should not be underlined (unless you speak German ;-). change to “should be emphasised”

line 137 “mixing state degree” -> sounds weird either use “degree of mixing” or “mixing state”

line 149f “can be found in for example, Poulain et al 2020” -> “can be found, for example, in Poulain et al 2020.

line 153: “Figure 1 demonstrates” -> it is not the Figure that does something. Better use “Figure 1 shows/depicts”

line 160: “within the diameter ranging from 5 to 800nm” -> “with a diameter range of 5 – 800 nm”

line 170f “respectively pass through” -> “respectively” cannot be used like that. This is also an example for a “;” monster sentence. Simply start a new sentence. “... monodisperse particle fraction. After the DMA, the flow was split to pass through a CPC [...] and a CCN counter [...]”

line 200 “was firstly corrected” -> was first corrected

line 203: “thus they are falsely selected in the DMA” -> they are selected in the absolute correct way. It is the assigned diameter that is incorrect. Simply remove this phrase.

line 204 “For this was corrected” -> “To correct for this, the fraction of multiple charged particles [...] was subtracted [...]”

line 216 “rather than an intermittent mutation” -> do you mean “rather than displaying (?) an intermittent mutation”?

line 235 “determined” -> determined feels a bit strong here. Maybe better “derived” since this is a approximation of the true k value?

line 278 “...gradually peaks in summer...” -> I do not know what “gradually peaks” means in this context

line 282f “the power-law and the error function” -> should be “a”.

line 285 “because of more parameters” -> “due to the higher number of parameters”.

line 298 “CCN number size distribution” -> missing “The”

line 382: What is meant by “aerosol cluster”?

line 415: “two categories of NCCN prediction approach” -> “approaches” or better “can be divided into two categories”

line 417 and later “category approach” -> only “category” without approach

line 444f “provide rough estimates on account of the pretty high RD” weird. RD is not causing the rough estimate it is the consequence. Better “provide rough estimate which is reflected in the high RD”

line 454: “...Figure 9 further evaluates the model...” It is not the Figure that evaluates the models.

line 458 “results are much uncertain” -> “the results have a high uncertainty”

line 466f “the prediction results remain a high uncertainty” -> ??? “the uncertainty of the prediction results remain high”???

line 475: “N<sub>CCN</sub> is overestimated at assuming a constant k” -> “when assuming”

line 478f: “the largest median overestimation reaches to 30%” -> no “to”

line 485f: “the 3<sup>rd</sup> scheme has better predictions on N<sub>CCN</sub>” -> “provides better predictions of N<sub>CCN</sub>”

line 510: “gradually changes” really? I would not call the big improvement from scheme 1 to 2 to 3 “gradual”. For the changes going from schemes 3-4-5, gradual is the correct term.

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