

Interactive comment on “Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China – Part 2: Size-resolved aerosol chemical composition, diurnal cycles, and externally mixed CCN-inactive soot particles” by D. Rose et al.

Anonymous Referee #3

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Overview

This paper discusses findings from a field campaign in China and is a companion paper to one published in 2010 in ACP. This paper extends the findings from the companion paper concerning Cloud Condensation Nuclei (CCN) analysis, adding chemical composition measurements from an Aerosol Mass Spectrometer, volatility measurements from a Volatility Tandem Differential Mobility Analyzer and optical properties from a

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nephelometer and a photoacoustic spectrometer. They estimate various hygroscopicity parameters (kappa) from these measurements and compare them to those that were measured with a CCN counter from the first paper.

This paper is a nice addition to the scientific literature concerning hygroscopicity parameters derived from CCN measurements and addresses relevant scientific questions. It is well within the scope of papers published by ACP. The methods and assumptions are clearly presented. There are sufficient references to the literature and previous work. This work merits publication after the following points are considered.

Major point

You refer to the soot particles in your title as “CCN-inactive soot particles”. And yet, in Section 3.2.1, you go to great lengths to determine a hygroscopicity parameter of the low volatility aerosol particles (kappa_lv). If it truly is CCN-inactive, there is no reason to determine a kappa value for this. By simply modifying your wording from CCN-inactive to something else, like “less CCN-active” or “less hygroscopic” versus more CCN-active or more hygroscopic, this may eliminate a lot of the confusion that threads through this work. For example, it was worded very nicely on page 26861 lines 6-7 “because kappa_t is the combination of the hygroscopicity of the CCN-active particles (kappa_a) and the much lower hygroscopicity of the lv-particles (kappa_lv).” Next, I will point out a few other suggestions along the same lines that were confusing but could be easily remedied.

I found parts of Section 3.2.1 confusing. With some slight changes in wording, it may become clearer. To try to illustrate my difficulty, page 26856 line 6 states “non-activated fraction may consist of fresh soot particles” and lines 17-18 state “non-activated particles jumps from following the lv-particle size distribution”, and page 26857 lines 7-8 “This is another indicator that the particles with a size of 270nm that did not activate at S =0.068% corresponded to the lv-particles at that size.” which says to me that “non-activated” particles do not activate, and that lv particles do not activate. You go to a lot

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of trouble to prove that these particles to not activate at 0.068% (figure 6, 7) but you should try to bring out more in this discussion that these particles DO activate at larger supersaturations and therefore DO have a hygroscopicity parameter worth calculating (i.e. non-zero). You might add something about this for example on page 26855 lines 23-25 and page 26856 lines 18-23.

In addition, I found the explanation of how you derived Equation (2) a little confusing. For example, why do you divide by “the number fraction of low volatility particles” (phi) in Eq (2)? At S=0.068%, from figure 7a, the relationship derived from the graph (the -1:1 line) is approximately $MAF=1-\phi$ at 270 nm. At higher supersaturations, MAF at 270 nm approaches 1 but what happens to phi? Is it possible to add a plot of phi vs D to an earlier plot in the paper, such as adding it to Figure 6 which is discussed in this section (Section 3.2.1)? Currently, it is included later on, in Figure 11, which has not been introduced during the Eq (2) discussion. If it's included, perhaps a comparison of phi vs D to figure 3 in the companion paper (Rose 2010) may help clarify this relationship shown in Eq (2). Whether or not phi vs D helps in this discussion, further explanation of where this equation (2) came from is required.

You carry on in the following section with distinctions like: page 26859 lines 5-6 “effective hygroscopicity of CCN-active (κ_a) and -inactive particles (κ_{lv}) on the assumption that $1-MAF_m$ comprises the fraction of CCN-inactive particles.” Also on page 26863 line 9 “fraction of the CCN-inactive lv particles”, in Table 1 (definition for κ_t), and in the conclusions on page 26864 line 10, page 26865 line 5 and line 8, you mention a “CCN-inactive” component. Please reword in these locations.

Minor points

Page 26843 Line 9: insert the word “the” before “AMS” so it reads “determined by the AMS”

Page 26847 Line 7: one or two sentences of explanation (including a reference if available) is needed for the TDMPS. What does “twin” refer to here? Who manufactured

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this instrument? Which instruments are used? Did you calibrate this instrument and if yes, what was the method? What are the uncertainties in these measurements? Are there references for its use in the past? All of the other instruments have their own section such as CCN, AMS, VTDMA, nephelometer/PAS but not the TDMPS.

Page 26847 Line 10: do you need the acronym “(IFT)” here? It is not mentioned again except in the acknowledgements where it is redefined

Page 26848 Lines 1-4: Combine the sentences “Details about the measurement procedure, calibration, and data analysis are described in Part 1 of this paper.” and “For a detailed description of the parameters see Rose et al. (2010a).” so that only one reference to the Rose et al. part 1 paper is needed.

Page 26849 Line 6: is this 50 nm referring to Dva or mobility equivalent diameter? I would subscript any mention of a mobility equivalent diameter to be something like Dm. For example, in Figure 1, is this Dm or Dva? If there is a subscript “m”, there will never be any question as to which Diameter it is referenced to.

Page 26850 Line 4: I would prefer capital letters for “LV” rather than “lv”, so that the first letter is easier to distinguish and the “L” is not confused with the capital letter “I” or the number “1”.

Page 26850 Line 18: was the time resolution 2 minutes for both instruments, the neph and PAS? If yes, change this to “; time resolution for both instruments was 2 min.”

Page 26851 Line 2: remind us here of the time interval for the CCN, such as “over the time interval of one CCN measurement (16 min).”

Page 26852 Line 2: add “Dm=” before 220 nm, then it should be clear for the rest of this section which diameter is referred to between Dm and Dva.

Page 26852 Line 3: I am not sure what “slightly above” refers to. For example, does it mean “slightly above the organic peak” or does it mean the “sulfate size distribution peaks at a mobility equivalent diameter (“Dm” if you like) that is slightly above 250 nm”?

Please clarify. If it is the latter, please provide the actual peak diameter and remove “slightly above”.

Page 26852 Lines 7 and 11: move “(Fig.1b)” from line 11 to line 7 after “(BBE)”.

Page 26852 Lines 27: You mention $\kappa=0.1$ for organics from Gunthe et al 2009, a study that took place in the Amazon (a rather different field campaign location than the current study in China). It might be nice to point out the range of κ values possible for organics from the literature, and thankfully you already cite many literature sources dealing with this topic on page 26844 line 24 to page 26845 line 7, as well as Petters and Kreidenweis, 2007 so just draw on those papers to provide this range of organic κ s.

Page 26853 Line 26: which supersaturations are included for the observed_kappa data determined from the CCN measurements in figure 3? Also add this information to the figure 3 caption.

Page 26854 Line 6: which supersaturations are included for the observed_kappa data determined from the CCN measurements in figure 4? Also add this information to the figure 4 caption.

Page 26854 Line 8: remove comma after “Note,”

Page 26854 Line 16: in brackets you have (NCCN,S) but it actually sounds like you are describing the methodology for determining the predicted CCN number concentration, which you label elsewhere as NCCN,S,p. I had thought that NCCN,S was observed from the CCN counter. Please clarify.

Page 26854 Line 19: point to Table 3 here where you first discuss the results, not just line 24 on this page.

Page 26854 Line 20: again, I would say here “between the NCCN,S,p predicted on the basis of $\kappa_{a,p}$ (Eq. 1)” to be clear on the differences between the NCCN,S and NCCN,S,p. Also, the way it is currently written, it implies that NCCN,S,p is included in

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Eq. 1, which it is not.

Page 26854 Line 24: change to “that the over-prediction of NCCN,S,p compared to NCCN,S becomes larger”, to be sure to draw attention to the correct column in the Table.

Page 26855 Line 3: in brackets, is Figs 2-3 referring to the current manuscript figures or figures in Rose et al 2010a? Please change to “(Figs. 2-3 in Rose et al., 2010a)”, if that is true.

Page 26855 Line 6-7: “Table 3 also shows the NCCN,S,p calculated for the same data set using a constant kappa of 0.3 (which is the campaign average value for kappa_t (Table 1)) as has been done similarly in Rose et al. (2010a).” should be reworded to something like “Table 3 also shows the NCCN,S,p calculated for the same data set using a constant kappa of 0.3 (which is the campaign average value for kappa_t (Table 1)), similar to the analysis performed by Rose et al. (2010a).”

Page 26855 Lines 11-12: change to “It confirms that kappa_a,p is not sufficient to predict NCCN,S,p well” if that is correct.

Page 26855 Line 26: this is the first time “MAF_f” has been introduced so please define it here or point to Table 1.

Page 26856 Lines 24 and 29: please remove “being the activated fraction measured at 270 nm;” from line 29 because MAF_m has already been defined on line 24.

Page 26856 Lines 27-28: reword to something like: “For all supersaturations, the measured CCN efficiency spectra had reached a plateau level at this size and thus did not increase further, which is referred to as the maximum activated fraction”

Page 26857 Line 3-4: I found this a bit confusing the way it is written: “as indicated by extrapolation of the lv-particle size distribution (Figs. 6 and 11).” Is this comparing the lv size distribution to CN size distribution? Please clarify. Also, you should either move figure 11 to figure 7 and introduce it at this point or remove “and 11” from the brackets,

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since we have not seen figure 11 yet, it has not been introduced yet, and it is presented out of order, i.e. it is presented prior to figures 7-10.

Page 26857 Line 6 and figure caption for figure 7: It is misleading to say it is a 1:1 plot when it is clearly not 1:1, it has a negative slope (-1). A decrease of 1 on the y axis gives an increase of 1 on the x axis, clearly not a 1:1 relationship. How about “Inversely proportional with a slope of approximately -1” or “-1:1 line”?

Page 26858 Lines 13-19 and figure 8b: If the S values were used from figure 8a to find points in figure 8b, why are there 4 points in 8a but only 3 in 8b? Explain here what happened to the fourth data point.

Page 26859 Line 1: reword to “In Fig. 9a, the kappa_t values predicted from equation 3 ($\kappa_t(ccn)$) are plotted versus the measured κ_t values.” Also, remind the reader where the “measured κ_t values” are from. With so many different variables and calculations, sometimes clarification is useful.

Page 26860 Line 10: Please change “In analogy to” to “Similar to” Section 3.3: How many days of data or how many data points are used to determine these diurnal cycles? Include this information in the figure caption or at the beginning of this section if possible.

Page 26861 Line 28-Page 28862 Line 2: Reword this as follows; “For the smaller particles measured at larger supersaturations (panel a), κ_a exhibited a clear minimum in the evening (20:00 to 22:00) and maximum values during day time, whereas for the larger particles measured at lower supersaturations (panel b) hardly any diurnal variation existed (less than $\pm 10\%$ variation of the mean κ_a).”

Page 26862 Line 10: change “maybe” to “may be”

Page 26862 Line 17: reword so it says “also exhibited” rather than “exhibited also”

Page 26862 Line 21: put the word “a” in front of “a little during the day”

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Page 26862 Line 23: why are you presenting figure 12k before presenting figures 12i and 12j? Is it possible to reorder the text or reorder the panels?

Page 26863 Lines 16 and 19: Why is panel “o” discussed before panel “n”? Swap panels?

Page 26865 Lines 23-28 and Page 26866 Lines 1-6: On page 26865, it seems you are outlining reasons why $\kappa=0.3$ has been used in the literature but it could be interpreted, since it is in your conclusions, to be conclusions from your study. My suggestion is to combine these 2 paragraphs (the last paragraph from page 26865 and first paragraph from page 26866) so that it is clear that the reason you discuss the literature value of $\kappa=0.3$ is so that you can discuss your tests to this value. Also, on Page 26865 line 23, it states “As discussed before” and I was wondering if “before” refers to “in the literature” or if it refers to “earlier in this manuscript”. If it is “earlier in this manuscript”, please indicate which section it is discussed. Lastly, on page 26865 line 25, it states “recent simulations”, which may be referring to findings from your paper but reading further, it seems these are from the literature. Please provide clarification to this paragraph, so that the reason why this paragraph exists is clear in the context of your work.

Tables

Table 1 page 26874: It would be nice if all values you refer to in this paper are included in this table. I found myself looking in Table 1 for reminders about what ϕ_{lv} or Sc or single scattering albedo were. Either include a complete list of terms used within this Table 1 or provide an appendix table with definitions of all terms used.

Table 2 page 26875: in the caption, please specify that this size range is mobility equivalent diameter or “ D_m ” if you like, so that it is clear it is not “ D_{va} ”.

Table 3 page 26876: reword beginning of caption “Arithmetic means for each data point in Figure 4 of the observed...”

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Figures

Figure 1 page 26878 and Figure 2 page 26879: is the diameter on the x-axis referring to mobility equivalent diameter or D_{va}? Please mention in caption or include a subscript on "D" to clarify.

Figure 3 page 26880: should it be "least squares", not "leasts"?

Figure 4 page 26881: remove comma after "Note," in the figure caption

Figure 5 page 26882: which supersaturation? Write "for all supersaturations" here somewhere if that is the case.

Figure 7 page 26884: should it be "least squares", not "leasts"? Also, please add the equations, correlation coefficients and number of data points for the 2 red curves in this plot, similar to what you did for Figures 3 and 9. Recall a minor point above (Page 26857 Line 6) about the 1:1 line reference that is misleading, please change this terminology to reflect the plot, perhaps "-1:1"?

Figure 8 page 26885: The first sentence in this caption is confusing, change to: "(a) CCN efficiencies plotted against supersaturation and (b) cumulative hygroscopicity Distribution, H(...), for lv-particles of 270 nm plotted against the hygroscopicity parameter, kappa."

Figure 9 page 26886: the subscripts are difficult to read on the axes labels. Also, should it be "least squares", not "leasts"?

Figure 12 page 26889: this figure has a lot of information crammed into it and it is difficult to read the axes. Also, you label each panel of the figure with (a), (b) etc but you are missing (l) between (k) and (m).

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