

***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.**

D. Rose et al.

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Response to Referee #3

We thank Referee #3 for the review and the constructive suggestions for improvement of our manuscript, which will be implemented upon revision. Detailed responses to the individual comments are given below.

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Referee comment:

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 (κ_{lv}). If it truly is CCN-inactive, there is no reason to determine a κ value for this. By simply modifying your wording from CCNinactive 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 κ_t is the combination of the hygroscopicity of the CCN-active particles (κ_a) and the much lower hygroscopicity of the lv-particles (κ_{lv}).”

Author response:

Many thanks for the suggestion. Our formulations were based on the fact that at a given supersaturation aerosol particles can either be activated as CCN (CCN-active) or they cannot be activated (CCN-inactive), depending on their size and hygroscopicity. In order to avoid confusion, we intend to reformulate the relevant text sections using the general term “weakly CCN-active” instead of the supersaturation-specific term “CCN-inactive”.

Referee comment:

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 “nonactivated” particles do not activate, and that lv particles do not activate. You go to a lot of trouble to prove that these particles do 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.

Author response:

We hope that the revised terminology introduced above (weakly CCN-active instead of CCN-inactive) will help to resolve and avoid further confusion. In addition, we will follow the referee’s suggestion and add a sentence explaining that the ~ 250 nm particles not activated at $S \leq 0.27\%$ were activated at higher supersaturations (P. 26855 L. 25): “As stated in Rose et al. (2010a), the CCN efficiency spectra observed at low supersaturations ($S \leq 0.27\%$) did not reach full activation, indicating the presence of externally mixed weakly CCN-active particles with much lower hygroscopicity. However, most of the ~ 250 nm particles that did not activate at $S \leq 0.27\%$ were activated at higher supersaturations.”

Referee comment:

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” (ϕ) 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 ϕ ? Is it possible to add a plot of ϕ 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 ϕ

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.

Author response:

A plot of ϕ_{lv} vs. D_{VT} is already given in Fig. 5 together with $1-MAF_f$ vs. D_a . From this and from Figs. 6 and 7 we concluded that the fraction of 270 nm particles that are not CCN-active at $S = 0.068\%$ correspond to the lv -particles at that size (P. 26857, L. 6-8). In Eq. (2) we want to derive the CCN spectrum of the 270 nm lv -particles, because we want to know the critical supersaturation, the point at which these particles finally activate as CCN. Similar to the derivation of the CCN spectrum for all aerosol particles, the CCN spectrum for the 270 nm lv -particles is calculated by dividing the number of activated lv -particles $((MAF_m - (1 - \phi_{lv})) \cdot CN_{270})$ by the number of all lv -particles at that size and supersaturation $(\phi_{lv} \cdot CN_{270})$. The number of CN at 270nm (CN_{270}) cancels out and thus Eq. (2) remains.

Referee comment:

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.

Author response:

We hope that the revised terminology introduced above (weakly CCN-active instead of CCN-inactive) will help to resolve and avoid inconsistencies.

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Referee comment:

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

Author response:

Thanks, we will do so.

Referee comment:

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 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.

Author response:

We will follow the referee’s suggestion and add a section on the TDMPS in our revised manuscript (Sect. 2.2.6).

Referee comment:

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

Author response:

Since we add another paragraph on the operation of the TDMPS, we will use the acronym “IfT” in another place of the revised manuscript.

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Referee comment:

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.

Author response:

We will change this paragraph to: “Key parameters derived from the measured CCN efficiency spectra are briefly explained and listed in Table 1. For a detailed description of the parameters and more details about the measurement procedure, calibration, and data analysis see Part 1 of this paper.”

Referee comment:

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 D_m . For example, in Figure 1, is this D_m or Dva? If there is a subscript “m”, there will never be any question as to which Diameter it is referenced to.

Author response:

Thanks for the suggestion. However, since all calculations and plots in our paper refer to mobility equivalent and not vacuum aerodynamic particle diameters (P. 26848, L. 25-28) we do not need and therefore would not like to specify the mobility equivalent particle diameter by introducing the subscript “m”. For better clarification, we will add the following statement on P. 26849, L. 4: “. . . and only the mobility equivalent diameter is referred to in the remainder of this paper.”

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Referee comment:

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”.

Author response:

Thanks for the suggestion. We will change “lv” to “LV”.

Referee comment:

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.”

Author response:

The time resolution was 2 min for the nephelometer and 10 s for the PAS. The PAS data was averaged for two minutes to match the time scale of the nephelometer. We will add this information in Sect. 2.2.5.

Referee comment:

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).”

Author response:

Thanks, we will include “(16 min)” on P. 26851, L. 2.

Referee comment:

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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.

Author response:

As stated in the above response we would not like to introduce the subscript “m”.

Referee comment:

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”.

Author response:

Thanks for pointing out this imprecision. We will change the sentence as follows: “Averaged over the campaign excluding the biomass burning event (Fig. 1a), the organic and the sulfate mass size distribution peaked at mobility equivalent diameters of around 220nm and 250 nm, respectively.”

Referee comment:

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

Author response:

Thanks, we will do so.

Referee comment:

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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.

Author response:

The characteristic range of κ for individual organic compounds goes from zero for insoluble species (Petters and Kreidenweis, 2007) to ~ 0.3 for small soluble molecules such as oxalic acid (Mikhailov et al., 2009). This information will be added in the revised manuscript.

Referee comment:

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

Author response:

As stated on P. 26849, L. 16-18, in the correlation analysis between AMS and CCN data, CCN efficiency spectra with $D_a - \sigma_a < 50\text{nm}$ were excluded, which includes all spectra measured at $S = 0.87\%$ and some measured at $S = 0.47\%$. Therefore it should be clear to the reader that the data points shown in Fig. 3 were measured at $S = 0.068\%$, 0.27% , and 0.47% . Also, Tabs. 3 and 4 list values for these three supersaturations.

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Referee comment:

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.

Author response:

As stated in the above response we do not see the need to include this information in the figure caption.

Referee comment:

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

Author response:

Thanks, we will do so.

Referee comment:

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.

Author response:

Thanks for pointing out the inconsistencies regarding N_CCN,S and N_CCN,S,p in this and also in some following comments. We will change N_CCN,S into N_CCN,S,p where necessary. The same we will do for kappa_a and kappa_a,p as well as for kappa_t and kappa_t,p. When we use the acronyms without the subscript “p” (e.g., kappa_a, kappa_t, N_CCN,S) we do not necessarily refer to the “observed/measured”

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values of these variables but to the general meaning, i.e., effective hygroscopicity parameter or CCN number concentration. For clarification in the revised manuscript, we will adjust the description of these acronyms in Tab. 1 and we will add the word “observed” in front of κ_a , κ_t , $N_{CCN,S}$ in those case where we really talk of the observed/measured values. When we talk of the predicted variables we will either use the acronym with subscript “p” or add the word “predicted” in front of the acronym without subscript “p”. That is, “ $N_{CCN,S,p}$ ” and “predicted $N_{CCN,S}$ ” are equivalent formulations.

Referee comment:

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

Author response:

Thanks, we will do so.

Referee comment:

Page 26854 Line 20: again, I would say here “between the $N_{CCN,S,p}$ predicted on the basis of $\kappa_{a,p}$ (Eq. 1)” to be clear on the differences between the $N_{CCN,S}$ and $N_{CCN,S,p}$. Also, the way it is currently written, it implies that $N_{CCN,S,p}$ is included in Eq. 1, which it is not.

Author response:

See response above.

Referee comment:

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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.

Author response:

Thanks, we will do so.

Referee comment:

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.

Author response:

Thanks for pointing out this mistake. It is referring to Figs. 2-3 in Rose et al. (2010). We will adjust the text accordingly.

Referee comment:

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).”

Author response:

Thanks, we will do so.

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Referee comment:

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.

Author response:

See response above.

Referee comment:

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

Author response:

Thanks, we will change it to “(1-MAF_f; Table 1)”.

Referee comment:

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.

Author response:

We will shorten P. 26856 L. 28 to P. 26857 L. 1 to “... but reached a plateau level (MAF_m; cf. Fig. 3 of Rose et al., 2010a).”

Referee comment:

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

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increase further, which is referred to as the maximum activated fraction”

Author response:

As stated in the above response we will rewrite the sentence.

Referee comment:

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, 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.

Author response:

We will change this sentence to: “. . . as seen from comparing the lv-particle size distribution with the CN size distribution (Fig. 6)”.

Referee comment:

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”?

Author response:

Thank you for pointing out this mistake. Indeed it is no 1:1 line. We will change it to “1:(-1) line”.

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Referee comment:

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.

Author response:

The fourth data point in Fig. 8a is at $S = 0.87\%$. A 270 nm particle with a critical supersaturation of 0.87% would have an infinite small kappa value that cannot be displayed in Fig. 8b.

Referee comment:

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 kappa_t values.” Also, remind the reader where the “measured kappa_t values” are from. With so many different variables and calculations, sometimes clarification is useful.

Author response:

Thanks for pointing out this imprecision. As stated above we will check the use of the variables kappa_t and kappa_t,p and correct them where necessary.

Referee comment:

Page 26860 Line 10: Please change “In analogy to” to “Similar to”

Author response:

Okay, we will do so.

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Referee comment:

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.

Author response:

There were 30 days of measurements that went into the calculation of the diurnal cycles. We will include this information in the first paragraph of Sect. 3.3: "... obtained from 30 days of CCN, AMS, VTDMA, and optical measurements."

Referee comment:

Page 26861 Line 28-Page 28862 Line 2: Reword this as follows; "For the smaller particles measured at larger supersaturations (panel a), kappa_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 kappa_a)."

Author response:

Okay, we will do so.

Referee comment:

Page 26862 Line 10: change "maybe" to "may be"

Author response:

Thanks, we will do so.

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Referee comment:

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

Author response:

Thanks, we will do so.

Referee comment:

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

Author response:

Thanks, we will do so.

Referee comment:

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?

Author response:

The order of the panels in Fig. 12 had been chosen very thoroughly and thus we want to reorder neither the text nor the panels. The order of the panels was chosen such that in the first place it is in accordance with text flow. However, on the other hand we found it helpful if parameters shown for both small and large particles are plotted next to each other. In some cases it was therefore not possible to keep the panels in chronological order.

Referee comment:

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

Author response:

Please see the above response.

Referee comment:

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.

Author response:

We will remove “as discussed before” on P. 26865 L. 23 but keep the rest of the paragraph as is. It should be obvious that the “recent simulations” refer to findings from the literature because we also give the reference at the end of this sentence.

Referee comment:

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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.

Author response:

Thanks for the suggestion; we will include a list with other used symbols in Table 1.

Referee comment:

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

Author response:

Thanks, we will change the sentence to “. . . integrated over a mobility equivalent size range of 50–850nm. . .”.

Referee comment:

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

Author response:

Thanks for the suggestion. As the data in Table 3 are only partly linked to Fig. 4 we prefer not to reword the caption.

Referee comment:

Figure 1 page 26878 and Figure 2 page 26879: is the diameter on the x-axis referring to

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mobility equivalent diameter or D_{va} ? Please mention in caption or include a subscript on “D” to clarify.

Author response:

As stated above we use only mobility equivalent diameters in this manuscript, so we do not see any need to explicitly mention it in the figure captions again.

Referee comment:

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

Author response:

Thanks, we will correct it.

Referee comment:

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

Author response:

Thanks, we will remove it.

Referee comment:

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

Author response:

Each data point of 1-MAF_f vs. D_a refers to another supersaturation. We will therefore change the sentence as follows: “. . .(1-MAF_f for all supersaturations, red

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symbols)...”.

Referee comment:

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”?

Author response:

Thanks, we will change it to “least squares” and we will add the equations, correlation coefficients and number of data points for the red curves in Fig. 7.

Referee comment:

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, $H(\dots)$, for lv-particles of 270 nm plotted against the hygroscopicity parameter, κ .”

Author response:

Thanks, we will change the sentence according to your suggestion.

Referee comment:

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

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Author response:

Thanks, we will correct to “least squares”. The axes labels will be increased in the revised manuscript version.

Referee comment:

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).

Author response:

For better readability, we intend to increase the panel size and the axes labels in the revised manuscript. We did not name any panel “panel (l)” in order to avoid confusions with the capital letter “l” or the number “1” in the text.

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