Review for "Spatial and temporal CCN variations in convection-permitting aerosol microphysics simulations in an idealised marine tropical domain"

Paper summary and recommendation:

This paper disentangles the contribution of different processes to the overall CCN variability detected over a domain the size of a conventional general circulation model (GCM) grid box for the case of a convective tropical marine boundary layer. The study is performed in a simplified idealised setup. Feedback pathways between aerosol concentrations and the environment via radiative or cloud microphyiscal interactions are ignored. Thereby, an attribution of different processes to CCN variability due to spatial and temporal variability of size and number of the 3 mixed-modes contributing to the CCN budget (Aitken, accumulation and coarse mode) is obtained. The authors show that CCN concentrations may vary up to a factor 3-8 throughout the simulation domain. Understanding the origins of this variability is an important step towards estimating the potential biases of aerosol-cloud interaction estimates obtained by GCMs, which do not resolve this variability. I therefore recommend this article for publication in Atmos. Chem. Phys. following minor revisions.

We thank the reviewer for their review and note they recognise the paper's value in providing information to explore potential biases in aerosol-cloud interaction estimates from lower spatial resolution GCMs.

Our replies to each of the reviewers' comments are provided below (coloured red) and, where changes to the manuscript have been made, these are highlighted in the track-changes version of the document provided.

Minor Comments – general:

• I believe that your aerosol concentrations are spun up from an entirely clean (i.e. Naero=0.0 cm-3) atmosphere. Please state this explicitly in the manuscript. I agree with the authors that this gives you the opportunity to disentangle the individual processes. However, this may be at least partially responsible for the high variability in CCN (800%) obtained after 12h of simulation following the period of intensive updrafts. If that is the case, context should be provided for the interpretation of this estimate. If you initialised a homogeneous profile of e.g. accumulation mode aerosol, would you still obtain such a high degree of variability of CCN following the intense updraft period? Please comment.
Yes, that's correct – the aerosol concentrations were initialized to zero at the start of the simulation. In the revised manuscript, we have added a sentence to state this explicitly (lines 12-13, page 5). We also agree that the period of intensive updrafts at around 4-7h of simulation is a likely causing an unusually high degree of variability. We do already note the unusual nature of this period in the Abstract (page 1, line 17) and have added "with intense wind-speed conditions" to further suggest the connection between those conditions and increased sea-spray emission.

• The phrase "strongly convective" period (or conditions) seems to refer to different things throughout the manuscript. Sometimes the phrase seems to be used to refer to the time period of intensive updrafts and strong horizontal winds and sometimes to periods of intense rain fall. Please define this term and use it consistently throughout the manuscript.
We have clarified use of this phrase in the manuscript (page 1, line 30; page 5, lines 30-31; page 7, lines 5-6; page 8, lines 10-11). Mostly, we refer to the "strongly convective period" as meaning the period when the dynamical conditions (updrafts and horizontal wind speeds) are intense.

• It has been shown (e.g. Textor et al, 2006: "Analysis and quantification of the diversities of aerosol life cycles within AeroCom") that different assumptions made in modeling the sea salt flux may yield vastly different estimates of sea salt emission fluxes. How sensitive do the authors think their results are to their implemented SS emission parameterisation? Please comment.

The simulations apply the Gong et al. (2003) sea-spray source function, which includes the behavior of the Monahan et al. (1986) parameterisation, with additional parameter to control emission of ultra-fine sea spray particles. The parameterisation was used by many of the global models in phase 1 of the AeroCom intercomparison, as analysed by Textor et al. (2006). As we also explain in our responses to the other reviewer, in Figure 5 of the recent paper by Salter et al., (2015), several different sea-spray source functions are presented in terms of their emission flux against wind speed, with the Gong being in the mid-range of the different parameterisations. We therefore believe our results are not sensitive to the particular sea-spray emission parameterisations and would be robust if a different emissions scheme were used.

Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global Biogeochem. Cycles., 14, 1097-1103, 2003.

Monahan, E. C., Spiel, D. E., and Davidson, K. L.: A model of marine aerosol generation via whitecaps and wave disruption. Oceanic Whitecaps. Edited by EC Monahan and G MacNiochaill, pp 167-193, D Reidel, Norwell, Mass, 1986.

Salter, M. E., Zieger, P., Acosta Navarro, J. C., Grythe, H., Kirkevåg, A., Rosati, B., Riipinen, I., and Nilsson, E. D.: An empirically derived inorganic sea spray source function incorporating sea surface temperature, Atmos. Chem. Phys., 15, 11047-11066, doi:10.5194/acp-15-11047-2015, 2015.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, I., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777-1813, doi:10.5194/acp-6-1777-2006, 2006.

Minor Comments – specific:

• P3L10: Please include reference Zubler et al (2011):"Simulation of dimming and brightening in Europe from 1958 to 2001 using a regional climate model", JGR, doi:10.1029/2010JD015396.

Done

• P3L10-L12: Two recent studies have investigated the impact of resolution on aerosol variability and aerosol-cloud interactions in regional climate models down to the kilometre scale for boundary layer clouds. These references should be added:

◦ Possner et al (2016): "The resolution dependence of cloud effects and ship-induced aerosol cloud interactions in marine stratocumulus", JGR, doi:10.1002/2015JD024685.

◦ Weigum et al (2016): "Effect of aerosol subgrid variability on aerosol optical depth and cloud condensation nuclei: implications for global aerosol modelling", ACP, doi:10.5194/acp-16-13619-2016.

Thanks -- we added citations to these references in the manuscript.

• P3L18: Please clarify complexity of aerosol treatment here, as there have been numerous studies investigating the sensitivity of marine deep and shallow convection to simplified aerosol treatments.
We are not sure what the reviewer refers to here. In the final paragraph of this section we do refer to the simulations applying an aerosol microphysics, and the particular type of aerosol dynamics scheme is clearly described in section 2.1, which follows immediately from this section. We therefore feel the level of detail given in this Introduction part of the manuscript is sufficient.

• P3L23: Please rephrase "to well characterize".
By "well characterize" we mean that the model will represent the dominant sources of CCN variability and therefore simulated CCN variations would be expected to be realistic. We therefore feel the word "well" is appropriate here. However, on reflection, perhaps that word does not need to be stated explicitly. We deleted it and also removed the 2nd instance of "influences" later in the sentence to improve the wording.

• P3L25: How do the authors determine the "realistic" level of variability? Please add references here.
We cite Yang et al. (2011) as a study that includes a similar level of model complexity.

Yang, Q., W. I. Gustafson Jr., Fast, J. D., Wang, H., Easter, R. C., Morrison, H., Lee, Y.-N., Chapman, E. G., Spak, S. N., and Mena-Carrasco, M. A.: Assessing regional scale predictions of aerosols, marine stratocumulus, and their interactions during VOCALS-REx using WRF-Chem, Atmos. Chem. Phys., 11, 11951-11975, doi:10.5194/acp-11-11951-2011, 2011.

• P4L30: Please rephrase "only a short demonstration simulation is here carried out".
Done

• P4L31: Please rephrase "carried on".
Done

• P5L11: Please rephrase "becomes precipitating", "becoming more intense".
Done

• P5L22: Please rephrase "associated cold pooling".
Done

• P6L24: By which criterion do you define your simulation to have fully spun up? Please clarify.
As discussed earlier in the manuscript, the model is spinning up over the simulation, including the period of convective instability at ~6h. Whilst we do not have specific criteria, we consider our focus on the last 12 hours of the simulation to be after the initial spin-up of the dynamics and primary aerosol (sea spray) in the model. We acknowledge that the secondary sulphate particles may still be spinning up, and we do discuss this clearly within the existing manuscript text.

• P7L20: The correspondence between patterns in highest particle concentrations and smaller particle sizes in Fig. 3 is not obvious to me in this particular figure. Please elaborate, or remove comment.
As this is not a key element of the study we decided to remove this comment.

• P7L31ff: The second half of the day is not only characterised by calmer wind conditions, but also by intense precipitation between 12 – 18 h. I believe that it should be mentioned here.
Yes – we agree. We now refer to this at this point as the reviewer suggest.

• P8L31: “adjusts to the very strong sea-salt emission and wet removal”. However precipitation only really intensifies much later than 8h after initialisation. Please comment on the role of wet removal during this period.
Even if the precipitation rate is less intense than later in the simulation, it is in average equal to 10 mm/h over the 5-8h period. So, the wet removal process becomes effective at this period of precipitation onset. We clarified this in the manuscript.

• P9L31 – P10L1: Remove sentence “The relative decrease in …”. You already stated that it is linear. *Done*

• P10L30: “… sea-salt aerosol are transported vertically by turbulent diffusion”, I would have thought that the convective updrafts would also contribute? Please comment, or adapt text.
You are right. We clarified the manuscript.

• P11L9: Please rephrase “wind speeds condition”.
Done

• P11L21ff: The authors state that CCN variations can be as large as factor 8. This number is obtained 12h after the simulation (Fig. 8). At this time the winds subside and precipitation builds up. So, how well does it characterise the CCN variability obtained during the period of intense updrafts? It may be helpful to include a box diagram for 6h after initialisation in Fig. 8.
We explain already that the processes mean we consider the CCN variability to be well characterized in the simulations. As above, this strongly convectively unstable period is not so representative of typical conditions and we therefore feel additional box diagram is not needed.

Furthermore the authors state that the CCN variability is large whilst the accumulation mode variability is smaller. This is confusing as I would assume most CCN to stem from the accumulation mode (see Fig 7.). Please clarify.

• Fig 1: For illustrative purposes the authors may consider adding a line of adiabatic parcel ascent.
We added a line representing the adiabatic parcel ascent and its specific levels (LCL, CCL and LFC).

• Fig 2: Please rephrase “mean total top cloud height” to “mean cloud top height”.
Done.

Please rephrase “rain accumulation” to “accumulated rain” or “accumulated precipitation”.
Done.

• Fig 4: For clarification it may help adding day and night markers for the sulfate chemistry.
As described in the section 2, there is no diurnal cycle in the model (page 4, line 30).

• Fig6 and Fig7: Does your aerosol scheme specify modal boundaries for the Aitken, accumulation and coarse mode? If so what are these? These could be added in the model description section.
The process of mode-merging is explained in Mann et al. (2010), and yes the scheme includes so-called “separation diameters” which determine at what point the particle size has grown large enough to be transferred to the adjacent larger mode. The values used are those as revised in Mann et al. (2012) to better capture size distributions simulated with the more complex sectional aerosol scheme. We feel the existing references are adequate here and think it is not necessary to re-state the values used explicitly.

Mann, G. W., Carslaw, K. S., Spracklen, D. V., Ridley, D. A., Manktelow, P. T., Chipperfield, M. P., Pickering, S. J., and Johnson, C. E.: Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model, Geosci. Model Dev., 3, 519-551, 2010.

Mann, G. W., Carslaw, K. S., Ridley, D. A., Spracklen, D. V., Pringle, K. J., Merikanto, J., Korhonen, H., Schwarz, J. P., Lee, L. A., Manktelow, P. T., Woodhouse, M. T., Schmidt, A., Breider, T. J., Emmerson, K. M., Reddington, C. L., Chipperfield, M. P., and Pickering, S. J.: Inter-comparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model, Atmos. Chem. Phys., 12, 4449-4476, 2012.

• Fig 7: What causes the large variability in radius for the accumulation mode particles up to 8h after initialisation? This is discussed in the text on P8L30ff, but I would have thought that the SS emission radius would be tighter constrained and that wet removal processes play a larger role later during the simulation (after 12h) as the RR peaks. Please clarify.

The large variability in radius of accumulation mode particles is caused by several processes, including those mentioned by the reviewer. The model size distribution responds to the different rapid changes during this high wind speed period that is generating strong sea-spray emissions. We therefore expect both emissions effects and removal effects to be influencing the behavior of the model. The different influences are complex and we feel our current qualitative discussion in the text is sufficient.