Review for "Parameterising Cloud Condensation Nuclei concentrations during HOPE"

General Comment:

The paper introduces a new empirical parameterisation for CCN activation developed for LES simulations over Germany, which may be applicable for a wider range of geographical locations and aerosol conditions. The suggested fit for the CCN parameterisation is a function of pressure and vertical velocity, which was obtained by fitting simulated CCN concentrations over Germany during the HOPE campaign period (26th March – 20th June 2013).
The simulated CCN concentrations were obtained with the COSMO-MUSCAT model. To compute the CCN activation, the aerosol number was first estimated assuming a fixed mean size and sigma for each considered aerosol species. From the obtained number size distribution of aerosol, the CCN concentrations were computed using the Abdul Razzak and Ghan (2000) activation scheme.
In the paper it is argued that the (spatial) median CCN concentrations at each pressure level are representative for the entire domain over a sufficiently short time period. Therefore curves of simulated median CCN concentrations obtained for a range of considered updraft vertical velocities were fitted.

As the authors state there is a need for computationally effective, yet realistic, CCN parameterisations that surpass fixed size-distribution assumptions, or vertically constant aerosol concentrations. However, I am not yet fully convinced by their approach. I have summarized my main concerns below, followed by minor issues and technical remarks. Based on these concerns, I recommend this article to be considered for publication after major revisions.

Major Comments:

My main concern regards the physical justification for the chosen functional of the fit. This is currently not addressed in the paper and needs to be included in a future version. In particular, what is your physical justification for dropping the dependence of CCN on temperature and humidity? I.e. having CCN=f(p, w) only and not CCN=f(p,w,T,qv)? Your current formulation would lead to the exact same CCN profiles in simulations performed under high-pressure conditions for a cold and wet as well as warm and dry atmosphere. Once a justification for CCN=f(p,w) is provided, the physical justification for the dependence of CCN~arctan(log(w)), i.e. the functional, should also be provided.

Given the chosen functional for the fit, the revised version should state the method of fitting and quantify the quality of the fit. The authors acknowledge that it is not good at low vertical velocities and low pressures (I guess there are not enough points here, to constrain the fit better?). Yet, no statistical measure of fit quality is shown. Depending on the method, simple error metrics like the R2, or RMSE should be provided. In addition it should be demonstrated that the curve is neither over- nor under fitted. This could be demonstrated by a chi-2 test. The table in the appendix certainly shows some fit parameters to be very poorly constrained (in sign and order of magnitude).

The justification for fitting the median CCN concentration at each pressure level is given in Fig. 5, where the first two panels show the CCN concentrations to be fairly constant over all latitudes and longitudes when vertically averaged. Does this also hold for each individual pressure level? In the eyes of the reviewer, this needs to be the case in order to justify Fig.6. If more variability is seen at some of the 32 considered pressure levels, one could perhaps consider fitting across the median+interquartile range instead?

Minor comments:

1) P2 L15-25: You first make the point that the more complex the aerosol scheme, the better the performance. In the following paragraph you state that a good parameterisation should be as simple as possible. I do not disagree with this discussion, but in its current form it seems like a contradiction.

2) Structuring in sections 2-4 could be maybe rethought and clarity increased
2 Aerosol data
3 Aerosol evaluation
4 Aerosol and CCN concentrations during HOPE

Maybe structure it as:
2 Aerosol simulations
3 Aerosol measurements
4 Aerosol and CCN concentrations during HOPE

The suggested headings, can be different of course. However, I think the structuring of the methods/results in this part of the paper could be improved.

3) P3L3: “This time period...”. State time period explicitly (is not yet motivated at this point of paper)

4) P3L1/6: Repetition of basic COSMO-MUSCAT configuration (COSMO+MUSCAT).

5) P3L16: How important are secondary organics in your domain for CCN? How big is the error of your simulated CCN due to the negligence of SOA?

6) P3L31ff: Should the presence of dust not be relevant for mixing state assumption?

7) P4L14: I would argue the temporal evolution of ammonium nitrate not to be well captured throughout the simulation. Please comment.

8) P4L25: Size distributions only really seem to match along a very small transect of size distribution. In other activation relevant regions (R>~35nm), the disagreement is quite large (log-scale). It looks to me that you underestimate the number concentration in many regions that are activation relevant.

9) P4L30ff: Following my concerns of point 8. It may help to provide some quantitative estimates here to support your argument. I.e. how low is the number concentration of the “large” particles (what is large?), or how high the difference in number concentration of the “small” particles (what is small?).

9) P5L16-L18: This is an interesting result. Do you have any explanation for why this is?

10) P6L22: What is “short time period”. Can you quantify this exactly?

11) P6: How much do you trust your quantification of table3 given your biases in the mass and number concentration estimates?

12) Fig 6: Looking at Fig. 5, the 30th of April CCN concentration roughly seems to correspond the mean concentration of the entire period. Would the agreement between the two panels be as good on say, the 6th of May, where the deviation from the mean CCN concentration is larger?

13) Fig. 8: To demonstrate that your fit works, would it not be better to split your dataset in 2 and to fit the first half of your dataset, arrive at your best fit and then compare your fit results to the simulation for the second half of your dataset (not included in the fit)? Or is it necessary to perform the fit on a daily basis? In this case would it not imply that you would have to refit each time before simulating a different case when using this parameterisation in an LES? Please comment.

Technical comments:

The manuscript contains a few typos and requires rephrasing in some instances. Some are listed here:

P1L20+others: “for use in models” needs rephrasing. Either “developed for models” or “to be used in models”

P1L24: “however, for the sake of...”. Maybe rephrase as: (Petters and Kreidenweis, 2007). Therefore, for the sake of simplicity...”.

P2L31: Typo “An essential ...”

P3L34: Typo “compisition”

P3L35: Rephrase “affect results strongly” → “affect the results strongly”

P3L35: Rephrase ”… are not sufficient only very close to the pollution sources”

Fig 3:
I) a), b), c) labels missing
II) I am not sure I am convinced by showing the dotted lines, when I do not know which species they correspond to?
III) Add the meaning of the grey shading in figure caption + choose stronger distinction in color for shading between observations and simulations.