Dear Editor,

The authors would like to thank you for handling the review of our manuscript. The authors would also like to thank the reviewers for providing fair and timely reviews of the manuscript, which will result in a much improved publication. Below is a list of comments raised by both reviews, the authors response, and any manuscript changes as a result. Please note, that all page and line numbers refer to the original submitted version of the manuscript.

1. Response to Reviewer 1

1.1 Major Comments

Reviewer Comment:
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.

Author Response:
There is a well established physical justification for choosing an arctan function to describe the activation spectrum shown in Figure 6. Low vertical velocities correspond to low supersaturations, as a result very few particles are activated. As the vertical velocity increases, an increasing number of particles are activated until the activated fraction approaches 1 at high vertical velocities. This relationship is an inherent attribute of the Abdul-Razzak et al. (1998) parameterisation, shown in their Figure 4. This shape is best described by the arctan function. The arctan function is very flexible, since by varying the coefficients it can describe many relationships from an almost straight line, to an almost step function.

It should also be noted, however, that this technique doesn't necessarily need a physical justification for the shape of the function. The new parameterisation is simply curve fitting, and the information about physical and chemical properties of aerosols are encapsulated in the Abdul-Razzak et al. (2000) parameterisation.

Now concerning the comment about dropping the temperature and moisture dependence. The Abdul-Razzak et al. (2000) parameterisation was implemented using the modelled temperature, moisture, and aerosol concentrations. The vertical velocity was kept as a free parameter, allowing the CCN concentrations to be evaluated at 40 different vertical velocities, as stated in P6 L24. Therefore, vertical velocity must be a dependent variable, as well as pressure for the vertical coordinate. Given that the CCN estimates, indicated

by the circles in Figure 8, were derived from the modelled temperature and moisture, the dependency of CCN on these parameters is already captured.

Furthermore, temperature varies very little at a given pressure level, and therefore CCN activation depends very little on these comparatively small variations. For example, mean CCN concentrations at 283.15 K on 26th March 2013 are 2.66×10^8 m^{-3}, and changing only the temperature to 273.15 K while holding everything else constant gives mean CCN concentrations of 2.80×10^8 m^{-3}. Also, dropping the dependency on moisture is insignificant, since the parameterisation should only be applied at or above water saturation.

If one chooses to simulate one of the days which has been explicitly parameterised, then one need not worry about the temperature and moisture dependence. However, if one wishes to use the mean profile provided, or any other profile, but for a different region or time period, then knowledge of the temperature and moisture conditions is required. For this reason, the domain mean surface temperature and the domain mean integrated specific humidity have been provided for each day, as well as the mean over the time period. The responsibility then falls on the user to choose an appropriate profile for their specific needs.

Manuscript Changes:
P7 L1: Added: 'In Figure 6, low vertical velocities correspond to low supersaturations, and as a result very few particles are activated. As the vertical velocity increases, an increasing number of particles are activated, until the activated fraction approaches 1 at high vertical velocities. This relationship is an inherent attribute of the Abdul-Razzak et al. (1998) parameterisation, and is best described by the arctan function.'

P7 L24: Added: 'Users must first decide which CCN profile suits the needs of their simulation, and select the appropriate fit parameters. The domain mean surface pressure, temperature and vertically integrated specific humidity given in Table 2 may assist in this regard. The mean CCN profile can be employed for longer simulations not wishing to include daily variability.'

Appendix: Table 2 added.

Reviewer Comment:
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).

Author Response:
The method used to derive the fits in non-linear least squares, where the data is first approximated by a some model, and the model parameters are refined through successive iterations which minimise the errors between data and the model.

The normalised RMSE (nRMSE) has been calculated in order to quantify the quality of the fit, and this has been added to the revised manuscript. The chi-2 test is not appropriate to test whether the parameterisation is under- or over-fitted, since the data are non-categorical. Instead, the residuals (ARG parameterisation – best fit function) have been analysed for each day. Below are two examples from the 30.04.2013 and 06.05.2016.

Here it can be seen that the residuals are largest in the lower troposphere. This could indicate that the parameterisation presented in the manuscript is, if anything, under fitted in the lowest levels. Since the residuals are small relative to the maximum concentrations, modifying the parameterisation to account for this variability would be an unnecessary over-complication. It would also need to be done on a case by case basis, since there is no systematic bias between the different days. In the upper levels, the residuals are smaller. This indicates that the current form of the parameterisation is appropriate to capture the main features in the vertical distribution of CCN.

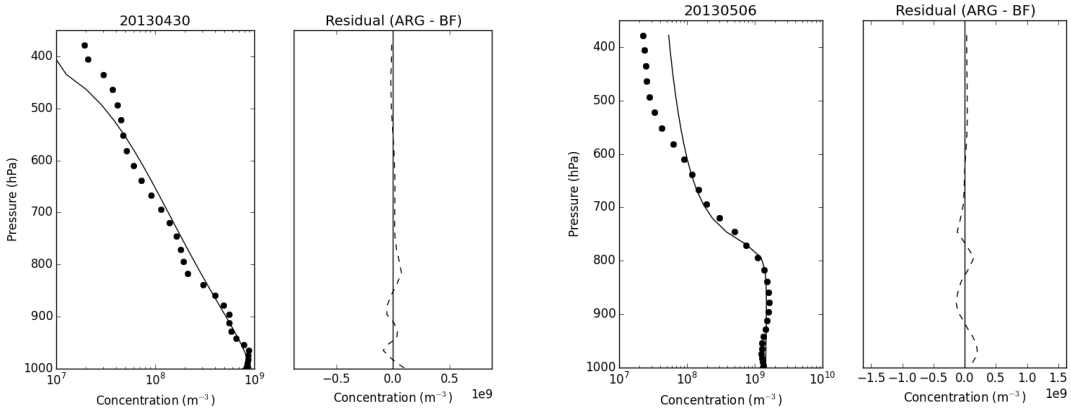

Figure 1: CCN concentrations from Abdul-Razzak et al. (2000) (dots) and from Equation (6) (solid line). The residual (Abdul-Razzak et al. (2000) – Equation (6)) is given by the dashed line for 30 April 2013 (left) and 06 May 2013 (right).

Manuscript Changes:
P7 L2: Added: 'The fitting was performed by means of a non-linear least squares method, where the data is first approximated by a model, and the model parameters are refined through successive iterations which minimise the errors between the data and model.'

P8 L19: Added: 'To further quantify the quality of the fit, the normalised root mean squared error (nRMSE) for each profile was computed, as is shown in Table 2. The

maximum nRMSE is 0.1930 for 03 June 2013, the minimum is 0.0127 for 01 April 2013, and the mean nRMSE over all days is 0.0555, which implies mean errors of 5.55%.
A qualitative inspection of the residuals, defined as Abdul-Razzak et al. (2000) - Equation (6), was also made (not shown). The residuals were largest in the lower troposphere, however still small relative to the maximum concentrations. This indicates that there is a small amount of variability in CCN concentrations that Equation (6) does not capture. In the upper levels, the residuals are much closer to zero, which indicates that the form of the parameterisation is appropriate to capture the main features in the vertical distribution of CCN.'

Reviewer Comment:
The justification for the fitting of the median CCN concentrations 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?

Author Response:
The figure below shows the latitudinal and longitudinal median CCN concentrations over all 32 pressure levels for 30 April 2013. For pressure levels close to the surface and extending to about 600 hPa, the CCN concentrations are still remarkably horizontally homogeneous. As indicated in Figure 5 of the manuscript, there are lower concentrations towards the north of the domain, where concentrations are slightly less than 40 % of the concentration in the middle of the domain. However, this covers a very small spatial area of less than 1 degree.

At higher levels around 400 hPa, there is larger variability. The concentrations to the north-west of the domain are about 6 - 7 times higher than the rest of the domain, which is due to the larger concentrations of hydrophilic sea salt aerosols in this region. The CCN concentrations at these higher levels are 2 orders of magnitude lower than those at the surface, and hence don't contribute much to the total CCN concentrations.

It is inevitable that one sees more variability in the individual pressure levels than in the mean, however the latitudinal and longitudinal median CCN concentrations are still relatively homogeneous. Therefore it is still appropriate to use the domain wide median CCN concentrations as representative of the whole domain.

Manuscript Changes:

P5 L26: Added: 'Even when individual pressure levels are considered, spatial variability in CCN concentrations remains low. The largest variability is in the north-west of the domain above 400 hPa, where concentrations are up to 7 times higher than the rest of the domain. CCN concentrations at 400 hPa are more than 2 orders of magnitude lower than near the surface, therefore the impact of this error is small.'

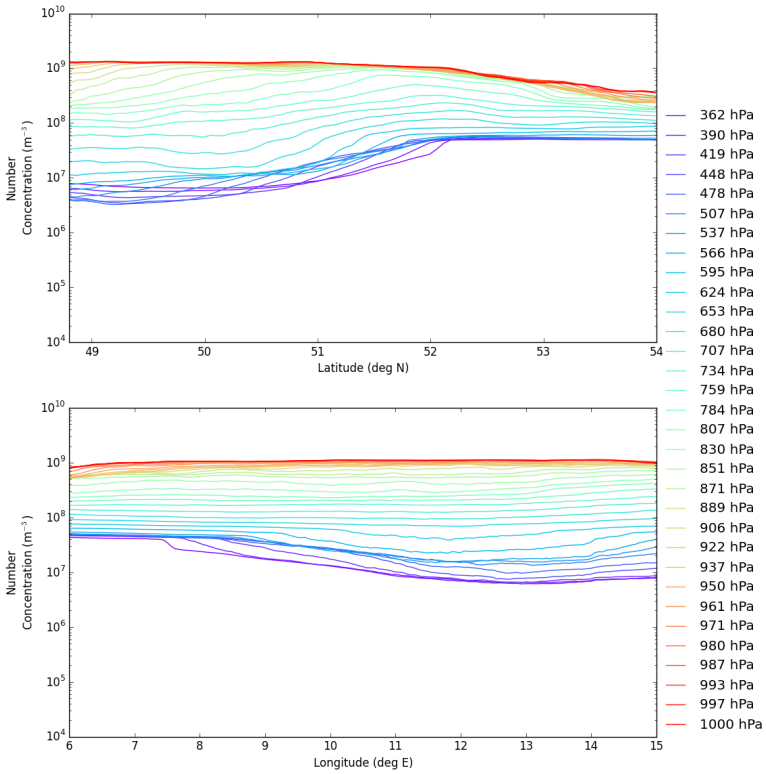

Figure 2: Longitudinal (top) and latitudinal (bottom) median CCN concentrations for 30 April 2013 at each pressure level.

1.2 Minor Comments

Reviewer Comment:
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.

Author Response:
Yes, in line 16 we make the point that a more complex aerosol scheme, the better the performance. Then in line 20 we make the point that an effective parameterisation should be as simple as possible. However, the reviewer neglected the second half of the sentence, which states '…, yet encompass all the governing factors affecting aerosol activation'.

The point is a good aerosol scheme should be complex enough to include all the governing factors affecting activation, but as simple as possible to facilitate efficient computation. There is no contradiction here when one reads the whole sentence.

Manuscript Changes:
None.

Reviewer Comment:
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 the part of the paper could be improved.

Author Response:
Thanks, the sections headings have been modified to increase clarity.

Manuscript Changes:
Section 2: Aerosol simulations
Section 3: Aerosol measurements and simulation evaluation
Section 4: Aerosol and CCN concentrations during HOPE

Reviewer Comment:
P3 L3: "This time period…". State time period explicitly (is not yet motivated at this point of paper).

Author Response:
The time period is stated explicitly later, in P4 L5.

Manuscript Changes:
P3 L3: 'This…' changed to 'The…'.

Reviewer Comment:
Repetition of basic COSMO-MUSCAT configuration (COSMO+MUSCAT).

Author Response:
Thanks.

Manuscript Changes:
P3 L6: removed ‘by COSMO-MUSCAT’.

Reviewer Comment:
P3 L16: 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?

Author Response:
The relative contribution of SOA to the total aerosol number (or mass) concentration is difficult to determine. SOAs have been found to be some fraction, often a large fraction, of total organic mass (Kroll et al., 2008). In our study, organic carbon has about an order of magnitude lower concentrations that the dominant aerosol species. Therefore, one can assume that SOA concentrations would be even lower than this.

Furthermore, a recent study (Zhao et al., 2016) found various SOAs have hygroscopicity parameters between 0.03 and 0.1. Whatever the concentration of SOAs are, their contribution to CCN would be very low due to the low hygroscopicity. Therefore, the errors in CCN associated with the negligence of SOA should also be very small.

Manuscript Changes:
P4 L18: Added: ‘Zhao et al. (2016) suggest secondary organic aerosols have hygroscopicity parameters between 0.03 and 0.1, less than the majority of aerosols considered in this study. Therefore, any underestimate in SOA concentration should not affect CCN concentrations significantly.’

Reviewer Comment:
Should the presence of dust not be relevant for mixing state assumption?

Author Response:
The results from Wang et al. (2010) suggest that the mixing state doesn’t effect CCN concentrations strongly, however they didn’t mention dust aerosols specifically. Sullivan et al. (2009) investigated the affect of chemical processing of dust and concluded that ‘it is incorrect to assume that all chemical reactions that process dust in the atmosphere produce soluble reaction products that increase the CCN ability of dust.’.

Furthermore, the dust concentrations in our simulations, from Figure 4, are nearly 5 orders of magnitude smaller than the dominant aerosol. Even if there was an enhancement in CCN activity of the dust due to the mixing state assumption, it would still be very small in comparison to the contribution of ammonium containing aerosols.

Manuscript Changes:
P3 L33: Added: 'Sullivan et al. (2009) show that not all chemical reactions that process atmospheric dust aerosols increase CCN activity.'

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

Author Response:
The variability is well captured, and the concentrations are well captured for most of the time period considered. There is an underestimate in the magnitude of the concentration of ammonium nitrate for the first few days of the time period. This was already acknowledged in the text in P4 L16. This has been more explicitly stated.

Manuscript Changes:
P4 L16: '…, where the model underestimates the observed aerosol species, particularly for ammonium nitrate, and to a lesser extent for ammonium sulfate and sea salt.'

Reviewer Comment:
P4 L25: 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.

Author Response:
This is already acknowledged in the manuscript in P4 L27 – P5 L4.

Manuscript Changes:
None.

Reviewer Comment:
P4 L30: Following my concerns of point 8. It may help to provide some quantitative estimates here to support your argument. I.e. how 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?).

Author Response:
The differences between the modelled and measured size distributions have now been quantified and discussed in the manuscript.

Manuscript Changes:
P4 L28: Added: 'For particles between about 10 – 30 nm, the model underestimates the number concentration by 1 – 2 orders of magnitude for the whole month. A smaller

discrepancy is seen for course mode aerosols, where the model suggests about half the concentrations given in the observations for 0.5 um particles.'

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

Author Response:
According to Abdul-Razzak et al. (1998), CCN activation is a complex function of temperature, pressure, moisture, vertical velocity, and aerosol concentration. As such, the variability in CCN concentration shouldn't be expected to exactly match the variability in aerosol concentrations. The relative contribution of each variable which affects aerosol activation has not been explicitly determined here, however it can be assumed that the temperature, pressure, moisture, and vertical velocity can act to smooth out the variability in aerosol concentrations at a given height (or temperature or pressure level). A quantification of this would be interesting, however it would be a digression from the aims of the manuscript. As this is not an essential part of the discussion, the authors wish to refrain from this type of speculation in the revised version of the manuscript.

Manuscript Changes:
None.

Reviewer Comment:
P6L22: What is "short time period". Can you quantify this exactly?

Author Response:
A short time period refers to one day.

Manuscript Changes:
P6 L22: Added: '… if short time periods of one day are considered.'

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

Author Response:
The biases in the modelled aerosol mass and number concentrations are, firstly, relatively small, and secondly, in the same direction. That is to say that the model always underestimates the mass concentrations when there's an error. Since Table 3 is comparing the relative contributions of aerosols to total CCN, the errors due to the biases should be small.

Manuscript Changes:

P6 L19. Added: 'Given that when there are errors in modelled aerosol mass concentrations the model is biased to lower values compared to observations, and Table 3 shows the relative contribution of modelled aerosols to total CCN, these errors should not affect these results strongly.'

Reviewer Comment:

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 concentrations is larger?

Author Response:

The shape of this activation spectrum is typical, as discussed in the first comment, and consistent between all simulation days considered here. The difference is in the aerosol concentrations at each level. The activation spectrum for the 6th of May, shown below, looks similar to the mean in Figure 6 of the manuscript, except the concentrations are higher. Here, the maximum concentrations near the surface reach 2.5 $\times 10^9$ m^{-3}, as opposed to 1.2 $\times 10^9$ m^{-3} for the whole time period. The shape of this activation spectrum has already been addressed in the revised version of the manuscript.

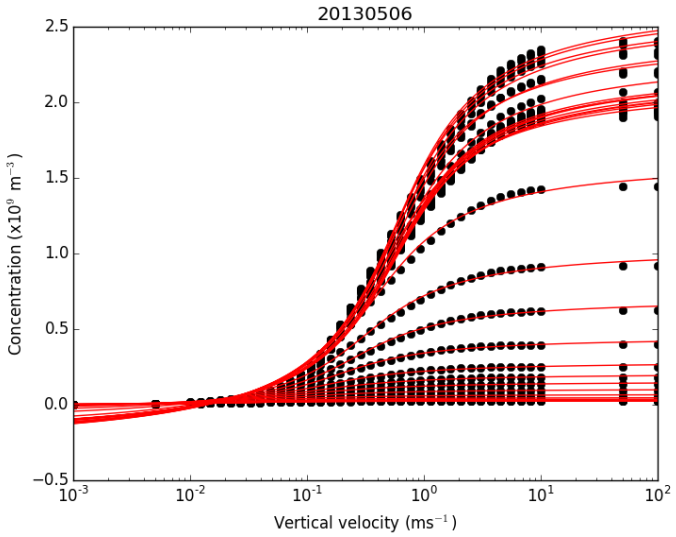

Figure 3: CCN activation spectrum for 6 May 2013. Black circles represent the model data, red lines are the best fit functions.

Manuscript Changes:

None.

Reviewer Comment:
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 preform 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 a LES? Please comment.

Author Response:
Regarding the quality of the fit, this has been quantitatively measured by the calculation of nRMSE and the residuals, which addressed the second comment. Also, Figure 9 of the manuscript indicates excellent agreement between the CCN concentrations from the parameterisation developed here, and from the Abdul-Razzak et al. (2000) parameterisation.

Regarding the second point, it is necessary to perform the fitting on a daily basis in order to ensure the CCN concentrations are more-or-less constant over the domain. The bottom panel of Figure 5 in the manuscript indicates there is up to an order of magnitude difference in the domain wide median CCN number concentrations on time scales of only a few days. This has already been noted in P5 L31 – 33.

The fitting of the CCN profiles has already been done for the whole time period of HOPE, and the fit parameters a1 – d4 of Equation (1) – (5) are given in the Appendix. It is not the users responsibility to do any fitting. Users of the new parameterisation must first decide which CCN profile(s) they wish to use, then simply read the fit parameters into the model. If days outside this time period are to be simulated, one could simulate aerosols and calculate CCN concentrations, and then fit them in a similar manner as demonstrated here. This has been clarified in the manuscript.

Manuscript Changes:
P7 L24: Added: 'Users must first decide which CCN profile suits the needs of their simulation, and select the appropriate fit parameters. The domain mean surface temperature and integrated liquid water given in Table 2 may assist in this regard, if users wish to apply them to days not parameterised. The mean CCN profile can be employed for longer simulations not wishing to include daily variability.'

1.3 Technical comments

Reviewer Comment:
P1L20+others: "for use in models" needs rephrasing. Either "developed for models" or "to be used in models"

Author Response:
Thanks.

Manuscript Changes:
P1L20: changed '…developed for use in models' to 'developed for models'.

Reviewer Comment:
P1L24: "however, for the sake of…". Maybe rephrase as: (Peters and Kreidenweis, 2007). Therefore, for the sake of simplicity…".

Author Response:
The word 'however' fits better than 'therefore', since the second half of the sentence doesn't follow directly from the first.

Manuscript Changes:
None.

Reviewer Comment:
P2L31: Type "An essential …"

Author Response:
Thanks.

Manuscript Changes:
P2 L31: changed 'A' to 'An'.

Reviewer Comment:
P3L34: Typo "compisition"

Author Response:
Thanks.

Manuscript Changes:
P3 L33: changed 'compisition' to 'composition'.

Reviewer Comment:
P3L35: Rephrase "affect results strongly" -> "affect the results strongly".

Author Response:
Thanks.

Manuscript Changes:
P3 L34: changed 'affect results strongly' to 'affect the results strongly'.

Reviewer Comment:
P3L35: Rephrase "… are not sufficient only very close to the pollution sources"

Author Response:
Thanks.

Manuscript Changes:
Changed '… are not sufficient only very close to the pollution sources' to '…are insufficient only very close to the pollution sources'.

2. Response to Reviewer 2

2.1 Major Comments

Reviewer Comment:
p.4, l. 15: What is the fraction of organic aerosol to total PM2.5? Does the underestimate of a factor of 2 mean that half of PM2.5 could be organic aerosol? How does this bias affect results, given that SOA has been shown to contribute significantly to CCN numbers?

Author Response:
This point was also raised by the first reviewer. The COSMO-MUSCAT model considers organic carbon, which judging from Figure 4 of the manuscript, has a concentration roughly an order of magnitude less than the total PM2.5. As stated in P4 L17, secondary organic aerosols (SOA) are not considered by the model, and as such, their number concentration can not be accurately estimated in our study. However one can assume that the concentrations should be less than those of organic carbon (Pandis et al, 1992).

A recent study (Zhao et al., 2016) found various SOAs have hygroscopicity parameters between 0.03 and 0.1, which is lower than most other aerosols considered in this study. These findings suggest SOAs should not contribute significantly to CCN production.

Manuscript Changes:
P4 L18: Added: 'Zhao et al. (2016) suggest secondary organic aerosols have hygroscopicity parameters between 0.03 and 0.1, less than the majority of aerosols considered in this study. Therefore, any underestimate in SOA concentration should not affect CCN concentrations significantly.'

Reviewer Comment:
In general, the CCN number concentration is a function of the supersaturation. The supersaturation is a function of the updraft velocity (source term) and the surface of the aerosol particles (sink term). If the total aerosol concentration is underestimated, I do not see how the CCN number can be calculated correctly in the model, only based on w.

Author Response:
The parameterisation developed here isn't only based on the vertical velocity. The physical and chemical properties of aerosols and the influence these have on CCN activation has already been included through the use of the Abdul-Razzak et al. (2000) parameterisation. The number concentration of aerosols is underestimated in the model, however the other dependencies on CCN activation are accounted for. This has already been explained in Section 5 of the manuscript.

Manuscript Changes:
None.

Reviewer Comment:
How well can w be constrained by the model? And by the observations? If CCN are parameterised such that they match observations, while there are uncertainties in both updraft velocity and total aerosol number (mass), it seems to be just an empirical fit where several effects might fortuitously cancel.

Author Response:
Observations are not used to derive the parameterisation. By 'observations', it is assumed that the reviewer means the CCN concentrations parameterised by applying Abdul-Razzak et al. (2000) to the modelled aerosol number concentrations.

The vertical velocity is held as a free parameter, and allowed to vary from 1×10^{-3} to 1×10^{2} m/s, and CCN concentrations are calculated at each vertical velocity. Therefore there are no errors associated with the vertical velocity. This has already been described in the Section 5 of the manuscript.

There is, however, an uncertainty in the modelled aerosol number concentration, as described in Section 3 of the manuscript. There are no other significant errors associated with the modelling of the aerosols, so it doesn't seem possible for errors to fortuitously cancel.

Manuscript Changes:
None.

Reviewer Comment:
At low updraft velocities, the supersaturation and therefore the CCN number concentration is the lowest. What is the reason for the largest uncertainty at these conditions? The scarcity of measurements, uncertainty in measurements or the fact that at low number concentrations of activated particles any bias translates into a larger error?

Author Response:
Measurements don't play a role in the parameterisation development. Modelled aerosol concentrations are used to estimate CCN concentrations, from which a series of best fit

functions are defined as a function of pressure and vertical velocity. This is the new parameterisation, as described in Section 5 of the manuscript.

The reason for the larger uncertainties at low vertical velocities is that this is simply where the differences between the best fit function, given by Equation (1), and the estimated CCN concentrations are largest. This has already been explained in P7 L3 – 5. This is noticeable in Figure 6 of the manuscript, where at vertical velocities between 1×10^{-3} and about 5×10^{-3}, the best fit function gives negative CCN number concentrations. In the model, and in reality, the CCN concentrations here would be simply zero.

These large errors are simply due to the best fit algorithm. The fit is not optimised for these parameter ranges, since large relative errors in low absolute concentrations contribute little to the total RMSE.

Manuscript Changes:
None.

Reviewer Comment:
What is the underlying reason that CCN concentrations are parameterised as a function of pressure? Is pressure only used here as a proxy of altitude.

Author Response:
The reason is two-fold. Firstly and most importantly, pressure is used as a proxy for altitude. Secondly, if height is used, then the model data shown in Figure 7 of the manuscript becomes more difficult to fit.

Manuscript Changes:
P7 L15. Added: 'In this series of equations, pressure is used as the vertical coordinate.'

2.2 Minor Comments

Reviewer Comment:
p. 1, l. 24: Reword. The hygroscopicity parameter implies some chemical composition, therefore composition is not neglected.

Author Response:
P1 L25 states '…, however for the sake of simplicity, chemical composition can be neglected. Segal and Khain (2006) state that…'. The reference to neglecting chemical composition relates to the work of Segal and Khain (2006), who don't use a hygroscopicity parameter, and hence make a simplified assumption about chemical composition. There is no contradiction here.

Manuscript Changes:
None.

Reviewer Comment:
p. 2, l. 6: Twomey's power law does not describe the size of aerosols but the total number of CCN.

Author Response:
Thanks for picking up on that. The paragraph has been modified accordingly.

Manuscript Changes:
P2 L5: 'Early parameterisations, including the seminal work of Twomey (1959), used a power law to describe the number of activated CCN. A power law can also been employed to describe the aerosol population, however this approach combined with simple expressions for the number of nucleated drops has drawbacks, since anomalously high droplet number concentrations can be produced.'

Reviewer Comment:
p. 3, l. 7: Table 2 is referenced before Table 1 (Is Table 1 referred to at all in the text?)

Author Response:
Table 1 should be referenced differently, since it's in the Appendix, and Table 2 should appear within the manuscript. Table 1 is, indeed, referenced in the text in P7 L23. The referencing will be different in the (hopefully) accepted and correctly typeset version of the manuscript.

Manuscript Changes:
None.

Reviewer Comment:
p. 3, l. 9: Reword. – Results of the study by Ghan et al. (2001) were used (or similar)

Author Response:
Thanks.

Manuscript Changes:
P3, L9: 'Ghan et al. (2001) was used…' changed to 'The hygroscopicity parameter for each aerosol species was taken from Ghan et al. (2001).'

Reviewer Comment:
p. 5, l. 6/7: The difference between ammonium sulfate and sulfate should be explained here (and not later in the manuscript). Is there no mechanism in the model that converts sulfate from nucleation into ammonium sulfate? Is this realistic?

Author Response:
In the model, SO2 is oxidized by OH to sulfate (which is supposed to be in the particle phase completely) and forms ammonium sulfate instantly if ammonium is available. Since the model does not treat particle numbers but masses only, this approach is realistic. In the higher levels, no ammonium is available, which is why the concentration of sulfate increases (see Fig. 4). This is described more in detail in Hinneburg et al. (2009) and Renner and Wolke (2010).

Manuscript Changes:
P 5, L 10: Added: 'Sulfate is the exception, with concentrations increasing with altitude. In the model, sulfate is formed by the oxidation of SO2 which instantly reacts to form ammonium sulfate, if ammonium is available (see Renner and Wolke (2010) for details). In the upper troposphere, less ammonium is available, which leads to the nucleation of sulfate particles.'

Reviewer Comment:
p. 6, l. 7: Related to my comment 2) above, the supersaturation is not only determined by the updraft velocity but also by the total aerosol surface – not only the aerosol species.

Author Response:
As mentioned above, the size of aerosols has been accounted for in this study. The dependence of supersaturation of aerosol size has been explicitly stated here.

Manuscript Changes:
P6 L7: Added: 'Of course this depends on the particular aerosol species, aerosol size, and thermodynamic conditions'

Reviewer Comment:
p.9, l. 2: Is it surprising that above the continents the anthropogenic fraction is greater that globally, given that the majority of surfaces are oceans?

Author Response:
No. The authors have already explained this in the manuscript P6 L17 – 19.

Manuscript Changes:
None.

1.3 Technical Comments

Reviewer Comment:
p. 1, l. 1: transportation -> transport

Author Response:
Thanks.

Manuscript Changes:
p. 1, l. 1: transportation -> transport

Reviewer Comment:
p. 2, l. 19: introduces -> introduce

Author Response:
Thanks.

Manuscript Changes:
p. 2, l. 19: 'introduces' changed to 'introduce'.

Reviewer Comment:
p. 2, l. 26 and 27: ICON-LES and HOPE should be defined here (and not in Section 2).

Author Response:
Thanks.

Manuscript Changes:
p. 2, l. 19: 'The parameterisation is developed for use in the large eddy simulation (LES) version of the ICOsahedral Nonhydrostatic (ICON) model, from modelled aerosol data during the HD(CP)^2 Observational Prototype Experiment (HOPE) campaign.'

Reviewer Comment:
p. 2, l. 31: A essential -> An essential

Author Response:
Thanks.

Manuscript Changes:
p. 2, l. 31: A essential -> An essential.

Reviewer Comment:
p. 3, l. 32: Black Carbon -> black carbon.

Author Response:
Thanks.

Manuscript Changes:
p. 3, l. 32: Black Carbon -> black carbon.

Reviewer Comment:
p. 3, l. 33: compisition -> composition.

Author Response:
Thanks.

Manuscript Changes:
p. 3, l. 33: compisition -> composition.

Reviewer Comment:
p. 3, l. 34: shouldn't -> should not

Author Response:
Thanks.

Manuscript Changes:
p. 3, l. 34: shouldn't -> should not.

Reviewer Comment:
p. 3, l. 34: shows -> show

Author Response:
Thanks.

Manuscript Changes:
p. 3, l. 34: shows -> show.

Reviewer Comment:
p. 4, l. 7: What does the T stand for in TDMPS?

Author Response:
T stands for Twin.

Manuscript Changes:
p. 5, l. 9: Added: '…using a twin differential mobility particle sizer (TDMPS)…'

Reviewer Comment:
p. 5, l. 9: aitken -> Aitken

Author Response:
Thanks.

Manuscript Changes:
p. 5, l. 9: aitken -> Aitken.

Reviewer Comment:
p. 5, l. 10: show -> shown

Author Response:
Thanks.

Manuscript Changes:
p. 5, l. 10: show -> shown.

Reviewer Comment:
p. 6, l. 27: dependence -> dependence

Author Response:
Thanks.

Manuscript Changes:
p. 6, l. 27: dependance -> dependence.

Reviewer Comment:
Figure 3: 1) The axes are very blurry; the exponents are hard to read; 2) The green dotted lines should be labelled; 3) add a), b) c) to the panels.

Author Response:
Thanks.

Manuscript Changes:
Figure 3 has been re-done.

3. Additional Changes

In the discussion version of the manuscript, the parameterisation is formulated as:

$$CCN(w) = A \times \arctan(B \times \log(w) + C) + D$$
$$A(P) = a1 \times \arctan(b1 \times P - c1) + d1$$

and so on for *B(P), C(P),* and *D(P).*

Notice that the sign of *C* and *c1 – c4* changes. This could lead to confusion during the implementation of the parameterisation. Therefore, the sign of the co-efficients for *c1 – c4* have been changed to positive.

4. Additional References

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