Referee Comment: *Estimation of Cloud Condensation Nuclei number concentrations and comparison to in situ and lidar observations during the HOPE experiments. This study compares CCN estimates from various methods. One method involves converting bulk mass measurements from both the COSMO MUSCAT model and from observations to CCN using assumed size distributions. CCN was estimated from these size distributions and compared to CCN observations (surface CCN, lidar, and in situ measurements on a helicopter) during the HOPE observational period in 2013. A second comparison involves comparing the 2013 estimates to estimates for the year 1985. Overall, I am unclear of the takeaways of this study, as their results seem to be primarily due to how the set up their methods, which are often not clearly stated or justified, and similar to a study that the authors were recently involved in (Hande et al. 2016). Generally, I found it somewhat difficult to assess the results and discussion and think the authors need to provide more details in several locations throughout the manuscript, including more explanations. This being said, I think there is a lot of interesting data and methods and I do think comparisons between models and parameterizations with observations can be useful. However, in the way this study was written and presented, I wasn't able to clearly discern this study's scientific contributions.*

Author Response: The authors would like to thank the anonymous Referee #2 for the constructive comments and suggestions made to improve the manuscript. According to the referee's comments, the authors have further improved the manuscript. All comments and changes in the manuscript are addressed below.

Referee Comment:

Major Concerns:

The calculation of the 1985 estimates

One of the main parts of this study is comparing results from 2013 to 1985. However, if I am understanding correctly, the manner in which the authors determine the 1985 values is by taking the 2013 simulated aerosol mass results and simply scaling them up by certain factors (Table 3). Then, the authors present the result that the CCN is higher for their 1985, but isn't this simply because they scaled up the mass concentrations in their methods. By simply scaling the 2013 model solution up and down, the authors may not be considering the different aerosol processes present in the model that may change with different emissions and concentrations within the model. For example, can the authors justify that the size distributions that are used to convert aerosol mass to the number should be the same in 2013 as in 1985. This may change their results significantly. I wonder if it would not make more sense to make assumptions about the emissions in 1985 and allows the model to run with those emissions, such that the processes are better represented with these higher concentrations are better represented. Also in Table 3, the authors do not explain where the scaling factors (3.9, 5.3, 2) come from? They also do not explain why they assume elemental carbon should be twice as high, since they themselves state there were no emission data to support this. More details are necessary to properly assess this study, especially since these are the primary methods in terms of estimating the 1985 concentrations.

Author Response: We are aware that the size distribution between the two scenarios 1985 and 2013 is likely different. We chose to assume the same size distribution because we found no justification to choose a particular different size distribution. Estimating a realistic dynamic size distribution requires explicit simulations of microphysical aerosol transformation processes, which was beyond the scope of this work, but is planned for a follow-up study. In the revised manuscript, we will discuss the shortcomings concerning the size distribution in a more extended manner.
A model run with emissions of 1985 was not conducted, since only limited emission information are available for this time period and also historical emission scenarios are affected by large uncertainties, in particular in the eastern European countries during Soviet times. Emissions would be needed spatially and temporally resolved, but only countrywide spatial average values of the annual sums were available. Scaling the emissions was not applied, since the spatial distribution of the modern emission data base does likely not fit to the 1985 industrial emitters. The emission

change is only known for the whole country, not distinguished by emission sectors or regions. Additionally, the mass-based model does not allow for the free evolution of the aerosol size distribution under altered emissions. Therefore, we chose to scale the concentrations of only that species for which we know how much the countrywide emissions have changed.
The derivation of the scaling factors between 2013 and 1985 is as follows:

- In the model, the formation of ammonium sulfate is described by pairing ammonia and sulfate until one of the two species is consumed completely.
  - 1985: High SO_2 concentrations lead to all NH_3 is going to ammonium sulfate.
  - 2013: Lower SO_2 concentrations leave a certain amount of ammonia unconsumed, which is then transformed to ammonium nitrate with the available nitrate.
- 2013: The SO_2 emissions of 0.41 Mt are completely transformed to ammonium sulfate ($(NH_4)_2SO_4$), which then consumes 0.22 Mt NH_3 to form 0.845 Mt ammonium sulfate.
- 1985: The emitted NH_3 (0.86 Mt) is completely transformed to ammonium sulfate, which results in 3.32 Mt ammonium sulfate. This process consumes 1.61 Mt SO_2, leaving 6.12 Mt SO_2 unconsumed.
- The scaling factor between 1985 and 2013 for ammonium sulfate is then calculated as 3.9 (3.32 Mt / 0.845 Mt).
- 50 % of the SO_2 left after ammonium sulfate formation (3.06 Mt) is transferred to sulfuric acid resulting in 4.68 Mt sulfuric acid. Since no further sulfate (sulfuric acid) is present in 2013, we calculate the 1985 sulfate concentration from the 2013 ammonium sulfate concentration. The ratio between the formed sulfate in 1985 (4.68 Mt) and the formed ammonium sulfate in 2013 (0.845 Mt) results in a scaling factor of 5.3.
- The doubling of the EC concentration is a rough estimate to address the strong decrease of EC emissions due to extensive reconstruction measures, replacements and shutdowns or state of the art emission control systems of power and industrial plants in the 1980s and 1990s. Since no detailed emission data are available, an exact treatment of EC is not possible. For this reason, a doubling of the EC mass concentration was chosen as a rough estimate. The EC concentration is not expected to significantly change the CCN concentration due to the generally low hygroscopicity of EC (see Table 1).

We will improve the description of the assumptions and calculations of the 1985 aerosol concentration estimates in the revised manuscript accordingly.

Referee Comment:

Unclear and unjustified methods

I appreciate that the authors' comprehensive study and comparing models to observations, which is a difficult task. However, throughout reading the manuscript, there were many instances where I either did not understand what the authors were doing and/or why they were doing it, which made it difficult to assess their results and conclusions. Although I have included some of these instances directly below, in general, I think the writing in the manuscript could be made much more clear and suggest the authors work on this.

Author Response: Thank you for the important comments. We will improve the revised manuscript accordingly.

Referee Comment: *P4, L15. The authors need to convert aerosol mass to a number size distribution, and therefore, must assume some size distribution shape. The authors end up choosing a one modal size distribution for each species, with parameters given in Table 1. Is it a good assumption to use one modal size distribution in this region? I am not particularly familiar with aerosol size distributions in this region, but I think the authors need to justify why using a one mode size distribution is valid and better justify the parameters chosen in Table 1, as opposed to just providing references. This assumption leads to one of their main results that they cannot CCN at higher*

supersaturation doesn't compare well, because they only used one mode at accumulation mode particle sizes.

Author Response: The assumptions made for the size distributions are explained, discussed and compared to measurements in more detail in Hande et al. (2016). They are based on AMS measurements of ambient concentration of the individual species (Poulain et al., 2011). A comparison of the estimated size distributions to observations can also be found in Hande et al. (2016). As can be seen in their Fig. 3c, the observed total aerosol size distribution at Melpitz is indeed bimodal with peaks at ~30 nm and ~100 nm diameter. The combined size distribution of the different species was found to match the observed size distribution well around 100 nm (i.e., the peak region of the largest mode), which is most relevant for estimating CCN. Nevertheless, the size distribution assumed here is used for the full domain. Applying a size-resolved aerosol transport model for a similar study would be a useful next step.

Referee Comment: *P8, L24-32: The authors clearly explain the process in which they estimate CCN. However, why do the authors not simply add up the CCN measured from from the different size selections to get the total CCN? The authors seem to currently convert the measured CCN to activated fraction, just to convert it back to CCN again? Can the authors clarify why they did this in this manner?*

Author Response: Thanks for pointing out this ambiguity in our description. In principal you are correct: It would be easier to just add up all measured CCN over the size range. However, if measuring monodisperse CCN one also has to account for multiple charged particles. These particles can have more than one electrical charge and as the particles are selected in the DMA by their mobility diameter, particles with more charges but larger diameter enter the CCNC at the same time. This effect influences the activation curve, because larger particles activate at lower supersaturation. Our multiple charge correction works in a way that the apparent diameter of doubly and triply charged particles is calculated and the actual fraction of those for each activation curve is calculated and subtracted from the activated fraction (AF). By doing so, we end up with the AF curve for single charged particles, which we use to derive the charge corrected CCN number.

Change in manuscript: To clarify this also in the text, we added a sentence (p8, l28):
The ratio between the CCN number and the total particle number as counted by the CPC (condensation nuclei, CN) gives the activated fraction (AF) of the particles. The AF was corrected for multiply charged particle up to three charges by subtracting their apparent fraction from the AF using the charge equilibrium (Wiedensohler, 1988). This multiple charge corrected AF is calculated for each particle diameter and results in a size dependent activation curve for each supersaturation.

Wiedensohler, A. (1988), An Approximation of the Bipolar Charge-Distribution for Particles in the Sub-Micron Size Range, J. Aerosol Sci., 19(3), 387-389.

Referee Comment: *P5, L20: "To achieve the maximum supersaturation (as a function of vertical velocity), accounting for particle growth before and after activation, the supersaturation balance is used." I do not understand this sentence and it seems to be important for the CCN calculation. What is the supersaturation balance? This paragraph in general, I think is very important, since it goes over how the estimated aerosol size distribution is converted to CCN, and therefore, I think it needs to be made much more clear how you are doing this. In its current form, I find it difficult to follow.*

Author Response: Overall, the paragraph was supposed to give only a little insight on the background of the widely used method derived by Abdul-Razzak et al. (1998) and Abdul-Razzak and Ghan (2000). We will improve this paragraph in the revised manuscript by making clearer what is actually used and avoiding unnecessary distracting information, but rather refer the reader to the two papers. The supersaturation balance was utilized by Abdul-Razzak et al. (1998) to determine the maximum supersaturation of an air parcel rising adiabatically at uniform speed and given aerosol type. However, the term itself (used by Abdul-Razzak and Ghan (2000)) is not widely used and is therefore perhaps distracting. The formulations were extended for multiple aerosol types by Abdul-

Razzak and Ghan (2000). Each aerosol particle can activate at a certain critical supersaturation. A formulation is derived, which describes how the critical radius of the smallest aerosol particle activated can be determined by the vertical velocity of an adiabatically rising air parcel: the critical supersaturation of the smallest activated particle is equal to the maximum supersaturation resulting from a uniform vertical velocity of an air parcel. Details of the derivation of the equations can be found in the papers of Abdul-Razzak et al. (1998) and Abdul-Razzak and Ghan (2000), to which it is referred to in the manuscript. Finally, in our study, the relation for the number of particles activated under given vertical velocity, aerosol composition and size distribution (Abdul-Razzak and Ghan (2000), Equation 13), is used.

Referee Comment: *P5, L25-27: I do not understand these sentences. What model are you referring to, and why would producing realistic supersaturations necessarily mean that you are also producing realistic CCN it also depends on if your aerosol number concentrations are realistic? Furthermore, why do the authors assume this model producing realistic supersaturations for stratiform clouds? I am confused by these sentences.*

Author Response: The word "model" was misleading. We refer to the activation parametrization, which was already evaluated by Hande et al. (2016). The calculated supersaturation here is depending on updraft velocity and the size distribution and hygroscopicity of the aerosol particles. Stratiform clouds are often horizontally more homogenic and can be better described with a grid scale vertical winds than convective clouds. Overall, the activation parameterization would also work for convective clouds if the updraft velocity would be known. We agree that this section was written confusingly and will improve it according to the given answer.

Referee Comment:

3: More explanations of the results

The authors present many comparisons, but often do not fully explain why there comparisons are the same or different. I have included some of these instances below.

P 11, L15-16: The authors state that nitrate is problematic to simulate (especially in spring). However, they just discussed how it was difficult to simulate in the fall. Therefore, why especially in the spring?

P11, L15-16: The authors state that nitrate is difficult to measure (especially in the fall). Why is nitrate more difficult to measure as compared to other species? Why especially in the fall?

Author Response: The terms "especially in spring" and "especially in fall" are misleading and not entirely correct. We will delete these terms in the revised manuscript and will clarify the uncertainties of both observations and modelling.
Today, the concentration of ammonium nitrate in agricultural regions is depending on the available ammonia. The ammonia emissions are in the short term uncertain since the exact timing of bringing out manure is usually not known. Hence, in particular the magnitude and timing of observed ammonium nitrate concentration peaks cannot be expected to match by applying ammonia emission data bases (time variation covers only the general seasonal cycle).
For the measurements the aerosol is collected on filters in High-Volume samplers without cooling. Since nitrate is volatile, warmer temperatures within the sampling and storage units than in the ambient air can lead to partial evaporation of nitrate from the filter. Therefore, the measured ammonium nitrate concentrations are a lower border of the actual ammonium nitrate concentrations.

Referee Comment: *P12, L3: The authors state that a "too large number of CCN could result from too many large particles". However, instead of speculating, the authors have the simulation data and the conversions to particle number size distributions and the observations to confirm whether this is the case and explain why.*

Author Response: Yes, this is true. We will re-write and extend the explanation in the revised manuscript. The model tends to underestimate the CCN concentrations, both for 0.2 and 0.3 %

supersaturation by a similar percentage (13 and 11 %). The particle concentrations are underestimated by the model as well, but this effect is more pronounced for smaller particles. The underestimation for particles larger 110 nm is about 10 % but 35 % for particles larger than 80 nm. This explains the larger difference in the ratio of $N\_CCN_{0.3\%}$ / N_CN > 80nm compared to $N\_CCN_{0.2\%}$ / N_CN > 110nm (Fig. 4).

Referee Comment: *P13, L5-7: This seems to be one main result of this manuscript, but it primarily based on data and results from a prior study (Hande et al., 2016). A possible contribution from this study would be to explain why their model is producing too many particles in the size range from 80 110nm?*

Author Response: The model underestimates both the particles larger than 80 nm and particles larger than 110 nm (see answer to previous comment) and therefore also particles between 80 nm and 110 nm. Since the applied chemistry transport model describes the mass concentrations of the different aerosol species, the calculated aerosol number size distribution, in particular its shape, depends on the assumed size distribution. The applied size distributions for the different species were compared to observations by Hande et al. (2016). The described underestimation is basically due to the utilized assumptions. These assumptions, however, have been proved by Hande et al. (2016) and were derived from observational data at the station Melpitz. Of course, we are aware that the chosen average size distribution is likely not representative for the whole domain and specific points in time. The present study, however, had the aim to provide estimates of the CCN concentrations in the 1980s using an offline method based on these assumptions. The respective paragraph was meant to discuss reasons for deviations between observation and modelled CCN concentrations. We will enhance the discussions at different parts of the manuscript and give more detailed information on the assumptions and known shortcomings. While there is always the possibility to design and conduct more sophisticated model experiments, we still believe that, despite the simplifying assumptions, our study provides valuable information on the CCN budget in the 1980s, which are of interest for the broader scientific community.

Referee Comment: *Figure 6 and P13 L13-20: One of the main conclusions of this study is that the vertical profiles compare well with the model and observations. However, all that is compared is an average over a month time period, and the authors conclude that it compares well because it is within a factor of 2 from the observations. Given the high temporal variability (seen in Figure 2), some temporal analysis should be considered here. Furthermore, it is difficult to read Figure 6 and see what the values and differences actually are.*

Author Response: A temporal analysis is included by displaying the temporal variability through the 0.25- and 0.75-quantiles of both the observations and the model. The plot does not show just the monthly mean, but mean of all measured profiles within the considered time periods. The model profiles were chosen for the exact times, for which the observations were available. We will improve Fig. 6 by adding the time periods and the summed observation time in the plot and the caption. Furthermore, we will make the plot wider and give vertical lines at the main ticks of the CCN number axis. The text will be revised by a better description of the data displayed in the Fig. 6.

Referee Comment: *Figure 7: One of the main results in this study is the shape of the CCN profile in Figure 7 and the differences between the model and observations. However, the authors don't really explain why it is different.*

Author Response: Figure 7 does not show observational data, but the comparison between the modelled CCN concentrations in 2013 and 1985. We assume, this comment refers to Figure 6. The specific reasons for the differences are not known. There are several reasons that likely all come to play, however their contribution to the overall deviation between model and observation is speculative: i) the observational methods have general uncertainties (e.g., factor 2-3 for the lidar retrieved CCN concentrations), therefore it is not possible to compare to a well-known truth ii) the

modelled aerosol concentrations are uncertain since the emissions are not known for short temporal and spatial scales, iii) the ambient aerosol size distribution is likely to deviate substantially from the applied mean size distribution, iv) the modelled boundary layer height is not as sharp as seen in the lidar observations (again for a number of potential reasons, which cannot be weighted by this study). We will enhance the discussion by giving potential reasons for the deviation between model and observation.

Referee Comment:

4: References

4A: Lack of reference

There is very little background included in this manuscript, many of which is likely very important to the authors results. On P2 L27, the authors state that comparison of modeled CCN to observations of CCN are sparse. However, since this is the entire point of this study, the authors should present the background literature here, as those results will likely be relevant to the results in this study.

Author Response: Agreed. In the revised manuscript we will add more references to previous studies that have contributed to evaluating model representations of CCN activation, including, e.g., Bégue et al., 2015; Spracklen et al, 2011; Watson-Parris et al., 2019; Nelson et al., 2016; Mann et al., 2012; Schmale et al., 2019; Almeida et al., 2014.

Begue, N., P. Tulet, J. Pelon, B. Aouizerats, A. Berger, and A. Schwarzenboeck (2015), Aerosol processing and CCN formation of an intense Saharan dust plume during the EUCAARI 2008 campaign, *Atmos Chem Phys*, *15*(6), 3497-3516.

Spracklen, D. V., K. S. Carslaw, U. Poschl, A. Rap, and P. M. Forster (2011), Global cloud condensation nuclei influenced by carbonaceous combustion aerosol, *Atmos Chem Phys*, *11*(17), 9067-9087.

Watson-Parris, D., N. Schutgens, C. Reddington, K. J. Pringle, D. T. Liu, J. D. Allan, H. Coe, K. S. Carslaw, and P. Stier (2019), In situ constraints on the vertical distribution of global aerosol, *Atmos Chem Phys*, *19*(18), 11765-11790.

Nelson, K. J., D. B. Mechem, and Y. L. Kogan (2016), Evaluation of Warm-Rain Microphysical Parameterizations in Mesoscale Simulations of the Cloudy Marine Boundary Layer, *Mon Weather Rev*, *144*(6), 2137-2154.

Mann, G. W., K. S. Carslaw, D. A. Ridley, D. V. Spracklen, K. J. Pringle, J. Merikanto, H. Korhonen, J. P. Schwarz, L. A. Lee, P. T. Manktelow, M. T. Woodhouse, A. Schmidt, T. J. Breider, K. M. Emmerson, C. L. Reddington, M. P. Chipperfield, and S. J. Pickering (2012), Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model, *Atmos Chem Phys*, *12*(10), 4449-4476.

Schmale, J., A. Baccarini, I. Thurnherr, S. Henning, A. Efraim, L. Regayre, C. Bolas, M. Hartmann, A. Welti, K. Lehtipalo, F. Aemisegger, C. Tatzelt, S. Landwehr, R.L. Modini, F. Tummon, J.S. Johnson, N. Harris, M. Schnaiter, A. Toffoli, M. Derkani, N. Bukowiecki, F. Stratmann, J. Dommen, U. Baltensperger, H. Wernli, D. Rosenfeld, M. Gysel-Beer, and K.S. Carslaw, 2019: Overview of the Antarctic Circumnavigation Expedition: Study of Preindustrial-like Aerosols and Their Climate Effects (ACE-SPACE). *Bull. Amer. Meteor. Soc.,* **100**, 2260–2283, https://doi.org/10.1175/BAMS-D-18-0187.1.

Almeida, G. P., J. Brito, C. A. Morales, M. F. Andrade, and P. Artaxo (2014), Measured and modelled cloud condensation nuclei (CCN) concentration in Sao Paulo, Brazil: the importance of aerosol size-resolved chemical composition on CCN concentration prediction, *Atmos Chem Phys*, *14*(14), 7559-7572.

Referee Comment: *4B: Inconsistent referencing practices.*

Furthermore, the authors have many inconsistent referencing practices. For example, they include references from some instruments and projects, and do not include references for others. The authors should take some time to include relevant references throughout the manuscript. Some specific examples are listed below:

P3 L19: What is GME?

Author Response: GME is the operational global icosahedralehexagonal gridpoint model, which was operationally used by the German Weather Service (DWD) previously to ICON. The reference Majewski et al. (2002) will be added to the manuscript. The abbreviation GME is a combination of its predecessor models GM (global model) and EM (a model for central Europe of DWD). Therefore, the abbreviation is not explained further and we refer to the reference.

Majewski, D., Liermann, D., Prohl, P., Ritter, B., Buchhold, M., Hanisch, T., Paul, G., Wergen, W., Baumgardner, J., 2002. The operational global icosahedralehexagonal gridpoint model GME: description and high-resolution tests. Monthly Weather Review 130, 319e338.

Referee Comment: *P7 L22 - P8 L2: Reference should be given for ACTRIS and EMEP.*

Author Response: The references ACTRIS (www.actris.eu) and Tørseth et al. (2012) will be added to the manuscript.

Tørseth, K., Aas, W., Breivik, K., Fjæraa, A.M., Fiebig, M., Hjellbrekke, A.G., Lund Myhre, C., Solberg, S., Yttri, K.E.: Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972–2009. Atmos. Chem. Phys. 12, 5447–5481 (2012).

Referee Comment: *P8 L3-4: References should be given to substantiate what HOPE is about.*

Author Response: The reference (Macke et al., 2017) for HOPE and its long name were given on page 2, line 31. The wrong brackets will be corrected in the revised manuscript.

Referee Comment: *P8 L9-12: References*

Author Response: The reference for ACTOS (Siebert et al., 2006) was given later in this paragraph and will be moved to this earlier mentioning of ACTOS.
The reference for LACROS (Bühl et al., 2013) will be added to the manuscript.

Johannes Bühl, Patric Seifert, Ulla Wandinger, Holger Baars, Thomas Kanitz, Jörg Schmidt, Alexander Myagkov, Ronny Engelmann, Annett Skupin, Birgit Heese, André Klepel, Dietrich Althausen, and Albert Ansmann "LACROS: the Leipzig Aerosol and Cloud Remote Observations System", Proc. SPIE 8890, Remote Sensing of Clouds and the Atmosphere XVIII; and Optics in Atmospheric Propagation and Adaptive Systems XVI, 889002 (17 October 2013); https://doi.org/10.1117/12.2030911.

Referee Comment: *P9, L1-2: Reference for this new instrument that the authors are using in the helicopter platform.*
If there is no reference, the authors should include more details about this instrument here, such as its specifications and where it was placed on the helicopter. The authors provide a lot of details in terms of ground measurements, but provide no details about the helicopter measurements.

Author Response: This mini CCN instrument is working by the same principle as the commercially available CCNC-100 by the DMT, which was used for the ground-based CCN measurements (Roberts and Nenes, 2005). It has also been used during other campaigns like in the CARRIBA campaign on Barbados (Wex et al, 2016).

Roberts, G. C., and A. Nenes (2005), A continuous-flow streamwise thermal-gradient CCN chamber for atmospheric measurements, *Aerosol Sci. Technol.*, *39*(3), 206-221.

Wex, H., et al. (2016), Aerosol arriving on the Caribbean island of Barbados: physical properties and origin, *Atmospheric Chemistry and Physics*, *16*(22), 14107-14130, doi:10.5194/acp-16-14107-2016.

Change in manuscript: We will clarify this in the text as follows (p8, l34):
"Within this study we use the vertically resolved in-situ data of the light weight mini cloud condensation nuclei counter (mCCNc, custom built by Gregory C. Roberts, working principal as in Roberts and Nenes, 2005), which has been applied successfully on ACTOS before (e.g., Wex et al. 2016)."

Referee Comment: *Table 2: Is the reference for this only personal communication? This seems rather weak, and given that this is the basis of the 1985 estimates, I think the authors should supply a better reference. Who is Kevin Hausman, for whom this personal communication was with?*

Author Response: The countrywide emission estimates were gained directly in personal communication from the German Environment Agency (Umweltbundesamt, UBA). Our contact person was Kevin Hausmann. From the time before 1990, no reliable emission records for especially the German Democratic Republic ("East Germany") were available. Therefore, the emissions for the 1980s with the assumed peak emission year 1985 needed to be estimated by Umweltbundesamt for the area of today's Germany based on data of the Federal Republic of Germany ("West Germany"). Kevin Hausmann provided the annual emission sums as spatial average over entire Germany. This information will be added to the references list in the revised version.

Referee Comment:

Other Comments:

Qualitative, subjective explanations: In general, the authors make several subjective statements instead of providing qualitative results that would be more useful to the reader. I have included a few instances below.

P11, L4: The authors state that the analyses shown in Figure 2 are in good agreement. However, this is qualitative and subjective. Can the authors provide quantitative, objective measures to describe their results here?

Author Response: This will be addressed in the revised manuscript.

Change in manuscript: We will change the sentence to (p.11, l.4):
On average (see Table 4), CCN concentrations from the direct measurements and the estimations based on chemical analytics agree within a factor of around 1.3. Taking the uncertainty due to assumptions in converting observed aerosol mass into number into account, the used CCN parameterization is concluded to work reasonably.

Referee Comment: *P11, L16: "results compare satisfactorily." This is subjective.*

Author Response: Apart from the two days discussed, on average the measured CCN concentrations are overestimated by 37 % for the fall period, and underestimated by 29 % for the spring period by the model. We will add this information in the revised manuscript.

Referee Comment: *P11, L20: The authors use a 1 to 1 scatter plot to compare their results, and state that the model data underestimates CCN compared to the other methods. Can the authors add more quantitative details here. Underestimate by how much? Can the authors possibly do least squares fits for their data in Figure 3 to accomplish this or some other method in order to provide some more concrete results?*

Author Response: We calculated linear regressions. For the spring period (Fig 3a), a slope of 0.59 and 1.47 was found for the comparison of CCNc observations to CCN number concentration derived from modelled and gravimetrically measured aerosol mass, respectively. For the fall period (Fig 3b), the slopes are 1.21 and 1.40, respectively. We will add this information to the plot and the text.

Referee Comment: *P12, L5: "Figures 2 and 3 show that the CCN number concentration is in similar agreement." Can the authors quantify this result.*

Author Response: This sentence refers to the two different supersaturations of 0.2 and 0.3 % (Figs. 2 and A1). We also calculated the average deviation between directly measured and calculated CCN number concentrations for 0.3 % supersaturation. Comparing the CCN number concentration derived from modelled aerosol mass to CCNc observations we found an underestimation of 25 % for the spring period and an overestimation of 32 % for the fall period. These are comparable to the values in Fig. 2 (29 % underestimation and 37 % overestimation, respectively).
We agree, that the respective paragraph in the manuscript is hard to understand. Therefore, we will revise the whole paragraph and also add quantitative information.

Referee Comment: *P13, L17-18: The authors state that the CCN number if overestimated by less than 50% and that this is quite well. This is subjective. Can the authors put this into context, in terms of how being 50% off would impact cloud processes or better explain why they think this is quite well?*

Author Response: The lidar-based CCN number concentration has a general uncertainty of a factor of 2-3 (Mamouri and Ansmann, 2016). Therefore, an overestimation (or better deviation) by 50 % is within this range. Furthermore, one cannot conclude that the model is 50 % off, but only that model and observation deviate from each other. Hence, the model estimate is in line with the observation. This is different for the comparison to the ACTOS profiles (which have an uncertainty of ~10 %). Here the model deviates from the observation outside its uncertainty range. The overestimation is 10-20 %. In the revised manuscript we will not use the qualitative term "quite well" and re-write this section including quantitative measures of the deviation between model and observation.

Referee Comment: *P13, L19: "decrease considerably." How much is considerably?*

Author response: The word "considerably" is just meant to qualitatively describe the obvious vertical decrease of the CCN number concentration from boundary layer to free troposphere seen in Fig. 6 (starting at around 1.3 km height upwards). The important finding is rather that the observations show a sharp decrease at the boundary layer height, whereas the modelled CCN number concentration decrease rather smoothly with height. Therefore, above the boundary layer the deviation between lidar-based and modelled CCN number concentrations increases to more than a factor of 2.

Referee Comment:

Unclear discussions

P12, L5-8: The authors first state that the results are in similar agreement, but then state that the differences are a factor of 2, and then state the difference is about 13%. I found these statements to be quite confusing. They must be comparing different things, but I wasn't sure what was being compared. I think the authors should be careful about making it very clear what they are referring to throughout their manuscript, since there are a lot of different data being presented in this study.

Author Response: The first statement, that the agreement is similar is referring to Fig 2 and A1. The main conclusion here is, that the comparison of modelled and observed CCN delivers similar results for both, 0.2 and 0.3% supersaturation.

The second statement refers to the peak deviation between CCN calculated from modelled and observed aerosol masses (Fig. 2 and A1 upper vs. lower panel). When averaging over the entire time period, a difference of about 13 % was found between the modeled CCN concentrations and the observed CCN concentrations with the CCNC. This can be seen from Table 4 as well (CCNC: $1.1*10^9\ m^{-3}$, model (2013): $9.4*10^8\ m^{-3}$).
We agree, that the paragraph should be revised to be less confusing and easier to read. Additionally, we will change Table 4 by moving the information of average measured CCN number concentration ($1.1*10^9\ m^{-3}$) from the table caption to the table content.

Referee Comment: *P13, L17-18: The authors state that the ccn number if overestimated by less than 50%, but that seems to only be the case of the lidar measurements but not the ACTOS measurements. Can the authors clarify? A much different picture is present in the ACTOS measurements, which are not really discussed.*

Author Response: The lidar-based CCN number concentration has a general uncertainty of a factor of 2-3. Therefore, an overestimation (or better deviation) by 50 % is within this range. Furthermore, one cannot conclude that the model is 50 % off, but only that model and observation deviate. Hence, the model estimate is in line with the observation. This is different for the comparison to the ACTOS profiles (which have an uncertainty of ~10 %). Here the model deviates from the observation outside its uncertainty range. The overestimation is 10-20 %. In the revised manuscript we will re-write this section including quantitative measures of the deviation between model and observation.

Referee Comment: *P11, L25-32: One large uncertainty in this analysis and comparison revolves around the size distributions being used in the model conversion from mass to number and whether these are representative for this site. As such, I think this should be included here.*

Author Response: It is true that the mass-to-number conversion is a large source for uncertainty. It was, however, developed from data at Melpitz and compared to observations at this site (see Hande et al., 2016). Therefore, it can be considered representative for this site. We will mention the mass-to-number conversion as source for uncertainty in this paragraph.

Referee Comment: *P14, L1-2. The authors state that there is increased variability in CCN number conc. In the free troposphere, which I think is based on the increased 25%-75% quartile range in Figure 5. However, the authors state this is "mainly an expression of the considerably increased detection uncertainty". However, the same trend is present in the model data, which does not have this detection uncertainty. Can the authority clarify why they believe this to be the case?*

Author Response: The decreasing trend of the median is observed in the model as well, but by far not as pronounced as in the lidar-based CCN observation. This observation method is more uncertain for small CCN concentrations as the lidar observations are close to detection limit. Therefore, the strongly increased variability is mainly due to the increased detection uncertainty and are thus statistically less relevant.

Referee Comment:

Additional Specific Comments

P11, L22-24: Since SOA is so important to this region, why wasn't it included in the COSMO MUSCAT simulations?

Author Response: By the time the simulations were conducted, the SOA module in COSMO-MUSCAT was under development and not sufficiently tested. Although, the available measurements at Melpitz point out a significant mass contribution of SOA in the PM1 (see Poulain et al., 2011), the calculated CCN concentration by applying the method described in our manuscript (applied size

distributions; activation according to Abdul-Razzak and Ghan, 2000; a factor ~4 lower κ-value of OC compared to ammonium sulfate or nitrate) to observed aerosol species masses was not determined by OC (see also Figure 2 c and d). Therefore, we decided not to include SOA. The statement given in the text was meant to express that SOA is important for the total organic mass. We will write this section clearer in the revised manuscript.

Changes in the manuscript: The difference in CCN from OC is partly due to the absence of secondary organic aerosol (SOA) in the model approach. SOA generally can contribute a large fraction to the total concentration of organic aerosol mass (Jimenez et al., 2009) and also at Melpitz SOA is known to comprise a major fraction of the PM1 aerosol (Poulain et al., 2011).

Referee Comment: *P2 L2: "For a realistic simulation of cloud adjustment..." What is cloud adjustment?*

Author Response: Cloud adjustment in the present context means the change of cloud macroscopic properties and dynamics due to perturbations of the aerosol. Whereas aerosol-cloud interaction describes the complex interplay between aerosols and clouds, cloud adjustments describe adjustments of clouds due to aerosol changes, such as the different aerosol loads between the 1980s and today over Europe. Examples of cloud adjustments are effects on cloud lifetime, cloud fraction or the timing and location of precipitation.

Change in manuscript: In the revised manuscript we changed the order and added context referring to "cloud adjustments" (p.2, l.2).
For a realistic simulation of microphysical aerosol-cloud-interactions and macroscopic cloud adjustment due to aerosol perturbations, a detailed representation of the aerosol in the models is required.

Referee Comment: *P2 L12-14: I do not understand what the authors are describing with these two sentences.*

Author Response: We wanted to express that in the COSMO-MUSCAT version described by Sudhakar et al. (2017), an interactive (two-way) online coupling was implemented. Where the aerosol transport part is frequently updated with meteorological fields and the meteorological driver can utilize the aerosol information in order to describe the activation of aerosol particles. The activation of CCN is treated as power laws and therefore, does not consider aerosol microphysical properties.

Change in manuscript: In the revised manuscript, the section will be changed (p.2, l.12):
This model version is online interactively coupled, making the activation of aerosol mass available for the two-moment scheme. However, the aerosol activation uses the bulk mass and does not explicitly consider aerosol microphysical properties.

Referee Comment: *P2 L32: Why 1985? Can the authors provide background information here as to why one would be interested in the year 1985?*

Author Response: The year 1985 is just the middle of the 1980s, which are considered to be the period with highest aerosol concentrations in Germany. Already in the late 1980s, emission reductions were applied. In the text, "1985" is equivalent to "peak aerosol scenario" or "1980s". We will include a clarifying statement in this section in the revised manuscript.

Referee Comment: *P3 L2: "This implies...". Can the authors clarify what this refers to? I am unclear and generally confused by this sentence?*

Author Response: "Implies" is the wrong word. Also, the sentence before was confusingly written.

Change in manuscript: The section will be re-written as follows (p.3, l.1):
The resulting CCN fields can be used in atmospheric models to analyze clouds and their radiation effects. For this purpose, CCN fields of variable degree of complexity can be generated, e.g., temporally and spatially constant CCN profiles, a 3D CCN field as a long-term average or even a 4D CCN field for temporally limited episodes.

Referee Comment: *P3 L4-9: The authors state they will be comparing 5 CCN estimates, but they seem to be estimates of different things. However, it is unclear why they are comparing these 5 items? As this is the introduction to the authors study, the authors should make this clear. What is the ultimate goal of this comparison and study?*

Author Response: Basically, with this list we aimed to give an overview of the CCN datasets from the different sources (model, chemical aerosol measurements, in-situ measurements) that were compared in this study. In short, the items are:

- CCN from the model for 2013 simulation
- CCN from the model for 1985 simulation ("peak aerosol scenario")
- CCN from chemical aerosol measurements
- ground-based in-situ CCN observation
- vertical CCN profiles from in-situ and remote sensing observations

We will re-structure this part of the text in the revised manuscript and make the description of the five datasets clearer.
The aim of the study is to provide estimates of the CCN concentrations in the 1980s using an offline method and compare to simulations and observations in the year 2013. The description of the study aims will be better summarised in the revised manuscript.

Referee Comment:

P3 L18: What is a composition cycle?

P3 L19: The authors state that the simulations will be reinitialized every 48 hours, and provide other details, without yet describing the basics of the simulation. Therefore, it is difficult to understand why a 48 initialization time is reasonable. It is suggested that the authors provide more details at the beginning of this section to make this section more readable.

Author Response: We agree, that the word "composition cycle" is unclear. COSMO and MUSCAT are coupled online, i.e., MUSCAT is updated with meteorological fields every time step. COSMO is initialized with coarser simulations and is updated only at the boundaries in order to make use of the higher resolved grid. This is also why COSMO is always run with a 24 h spin-up before the coupling to MUSCAT is switched on. After the spin-up period, COSMO and MUSCAT run coupled for another 48 h. In order to keep modelled meteorological fields close to the real atmosphere in these hindcast applications, we re-initialize the simulation cycle every 48 h, i.e. after 72 h for COSMO. We will improve this description in the revised manuscript.

Referee Comment: *P4 L8: Why do the authors only use model data up to 8km? The authors should provide a reason to justify or explain why they are not using the full model data?*

Author Response: The model simulations use more vertical levels (50) up to a height of 22 km. For the analysis, only the lowest 32 layers (8 km) were saved since we were mainly interested in the lower troposphere. We agree that the information is misleading and we will delete the sentence in the revised manuscript, and instead give the actual number of vertical levels in the simulation.

Referee Comment: *Figure 1: Why is only Melpitz shown, when Julich is also mentioned? Were measurements taken at the different city, Julich? I was confused about this throughout the manuscript.*

Author Response: In the present study, no comparisons for the site at Jülich were conducted. However, COSMO-MUSCAT was applied for simulations during the time period of the campaign taking place at Jülich in order to provide CCN data for this campaign and subsequent ICON-LES applications. For the comparisons in this study, we took data solely from the site in Melpitz. This was not clearly written in text and will be improved in the revised manuscript by using the terms "spring" and "fall period" instead of HOPE Jülich and HOPE Melpitz.

Referee Comment: *P4 L2: "in order to be considered" for what?*

Author Response: We mean that information on aerosol and CCN needs to be prescribed in order to be used for ICON-LES simulations, e.g., to alter cloud properties.

Referee Comment: *P4 L3: The authors state the model simulations were run. Are these ICON LES simulations, which were just mentioned in the previous sentence or COSMO MUSCAT simulations? I think it is the COSMO MUSCAT simulations, but can the authors make this more clear?*

Author Response: Yes, we used COSMO-MUSCAT. The calculated CCN fields were provided for ICON-LES simulations within the framework of the HD(CP)^2 project. We will make this clearer in the revised manuscript.

Referee Comment: *P4 L17-18. It seems that the authors imply that total mass concentrations is converted to total number concentrations before the log normal size distribution parameters are set? However, these size distribution would be necessary for the initial conversion to mass to number. So I am ultimately confused by what the authors are actually doing?*

Author Response: The process of activation is applied in two steps. First the aerosol particle number size distribution is calculated. Then these size distributions are used in the activation parametrization. We will write this in a clearer manner in the revised manuscript.

Referee Comment: *P5 L7: Which of the values are not according to Hande et al. (2016) and why?*

Author Response: The value of kappa for sulfuric acid was changed. In Hande et al (2016), a kappa value of 0.236 was assumed, which is far too low. Petters and Kreidenweis (2007) state mean values of kappa of 1.19 (growth factor derived) and 0.9 (CCN derived) for sulfuric acid. Based on these values, a kappa of 1 was chosen for this study.

Referee Comment: *P5, L14: The authors mentioned that ammonium sulfate has a kappa of 0.6, but then use 0.51 in their table. Why?*

Author Response: What was meant is that kappa of ammonium sulfate is around 0.6. In Petters and Kreidenweis, there are values from 0.33 up to 0.72 for the growth factor derived kappa and 0.61 for the CCN derived kappa. For consistency, we decided to use the same value as in the previous paper Hande et al. (2016) (taken from Ghan et al., 2001).

Referee Comment: *P5, L33-34 this should be included in the introduction.*

Author Response: We agree and will add this information in the introduction.

Referee Comment: *P5, L7: The authors reference Table 1, but mean to reference Table 2.*

Author Response: The sentence is about the parameters of the mass-to-number conversion and therefore correctly references Table 1.

Referee Comment: *Table 4: How were the measured aerosol mass concentrations obtained? Can the authors include this information in the caption, similar to how they mention that the modeled concentrations came from the COSMO MUSCAT simulations.*

Author Response: Yes, we will add according information about the gravimetrical measurements in the table caption.

Change in manuscript: Table 4. Average CCN number concentration (m^{-3}) and average contribution (%) of the considered species to the total CCN number concentration at ground level for a supersaturation of 0.2% at the HOPE site Melpitz for the 2013 campaigns and the corresponding period in 1985. The values were calculated from aerosol mass concentrations modeled with COSMO-MUSCAT and from aerosol mass concentrations observed by gravimetrical measurements. For comparison, the average value measured by the CCNC was $1.1*10^9$ m^{-3}.

Referee Comment: *P10, L16-19: The authors discuss why the measured dust was larger than the modeled dust. Can the authors make these statements more clear. For example, why is it OK to assume the difference in total mass from the measured species must be from dust?*

Author Response: In the gravimetrical measurements, different species, such as sulfate, nitrate, organic and black carbon, can be detected. The undetectable rest of the mass is generally accounted as "other" dust, i.e. just particulate mass which cannot be further speciated with standard methods. The model does only consider mineral dust from deserts. Additional primary dust sources, such as road dust, are not considered. Therefore, the model tends to underestimate the observed dust concentrations. Overall, the modeled and observed dust contributes only little to the total CCN concentration (see Fig. 2).

Referee Comment: *P11, L8: The authors state the nitrate was overestimated by a factor of 2. Can the authors present this information more clearly, possible on the same figure to make this more clear.*

Author Response: The statement refers only to the 3-day period (day of the year 255 – 257), which show a strong overestimation of the ammonium nitrate mass, and therefore CCN from ammonium nitrate (Fig. 2b and 2d). The difference between modelled CCN and CCN from chemical measurements of about a factor of two during those days can clearly be seen (Fig. 2b and 2d). Also, it can be seen that during those days, ammonium nitrate contributes most to the CCN in both observation and model. As for the CCN, the overestimation of ammonium nitrate is about a factor of 2 (CNN from ammonium nitrate: 1.0-1.5 x 10^9 m^{-3} for the gravimetrical measurements and 1.5-3 x 10^9 m^{-3} for the modelled CCN).

Referee Comment: *P11, L8-16: A majority of the discussion is focused on a 3 day period, which is very difficult to see in Figure 2. Can the authors create a new figure that zooms in on this period, to allow the reader to more easily follow along in the figure.*

Author Response: We did not intend to focus much on this period. However, we found it necessary to discuss this interesting, yet not important, feature. We will extend the general discussion on the findings around displayed in Fig. 2.

Referee Comment: *Figure 6: Why isn't the fall period shown here?*

Author Response: The fall period is shown in Fig. 6b. To make it clearer, we will add this information also in the figure itself and not only in the figure caption.

Referee Comment: *P14, L8. The authors state that for this calculation a vertical velocity of 1 m/s is used. I don't understand what this is referring too.*

Author Response: In contrast to the previous analysis in the manuscript that compared CCN at a fixed supersaturation to observations, for the last discussion part we calculated the CCN at a fixed vertical velocity (1 m s^{-1}). Following Abdul-Razzak and Ghan (2000), the maximum supersaturation can be calculated assuming an air parcel rising adiabatically with a given vertical velocity. This maximum supersaturation defines the critical radius of the given size distribution and therefore the number of particles activated to CCN. The maximum supersaturation, hence, depends on the particle composition and the number size distribution. The chosen value of 1 m s^{-1} serves as an example in order to compare the present day and 1980s scenario. In a further model development step, the aerosol composition could now be used to alter cloud microphysical properties under the given modeled grid or sub-grid scale vertical wind velocities.

Referee Comment: *Figure 7: What supersaturation is used for this analysis?*

Author Response: Fig. 7 shows the CCN number concentration for an updraft velocity of 1 m s^{-1}. This is mentioned in the text (p.14, l.8). We will add this information also in the figure caption. The supersaturation therefore depends on the aerosol chemical composition of the aerosol and the number size distribution and varies spatially and temporally (see also answer to previous comment).

Referee Comment: *Can the authors changes their units to cm^{-3} from m^{-3} , such that it is easier to comprehend the values presented?*

Author Response: Thank you for this remark. "cm^{-3}" are widely used, but we prefer SI units, which is also according to ACP guidelines.