Referee Comment: *This paper presents estimation of CCN number concentrations based on 1) simulated mass concentrations of major particle-phase species using COSMO-MUSCAT and 2) measured mass concentrations of major particle-phase species obtained during HOPE campaign in 2013. The ground-level model simulations and measurements were used to estimate the vertical profile of CCN number concentrations based on parameterizations reported in previous studies assuming log-normal number-size distribution of externally mixed particles.*

The estimated vertical profiles of CCN were compared with ground-based and airborne measurements during HOPE. In-situ CCN measurements were performed at the ground and on a helicopter. In addition, vertical CCN concentrations were estimated using lidar measurements. The vertical profile estimation agreed with observation near the ground but differed significantly at higher altitudes (Figure 6). The model was applied to peak aerosol scenario in Germany in 1985 to evaluate the anthropogenic impacts on CCN concentrations. No observation was available in 1985. They showed that higher anthropogenic emissions in 1985 resulted in higher CCN numbers compared to 2013 represented by the scaling factor (SF) > 1 (Figure 7).

I believe that the manuscript is technically sound and will be of interest to readers. However, I found the manuscript somewhat confusing and missing some details. I support publication after authors address the following specific comments.

Author Response: The authors would like to thank the anonymous Referee #1 for the consideration of the manuscript to ACP and for the constructive comments and suggestions made to improve the manuscript. According to the referee's comments, the authors have revised the manuscript. All comments and changes in the manuscript are addressed in the following.

Referee Comment: *1. A significant limitation of the CCN estimation appears to be that it neglects aerosol microphysics. If I understood correctly, the CCN prediction is linearly proportional to mass concentration since the size distribution is fixed. A large body of literature has shown that dependence of CCN number on emission is highly non-linear especially when microphysics of the nucleation and the growth of sub-CCN size particles into CCN size range are considered. For instance, Pierce and Adams (2009) showed that when primary emissions rates are varied by a factor of 3, tropospheric average CCN (at 0.2% supersaturation) only varied by 17% because higher primary emissions results in higher condensation and coagulation sinks. Although I recognize that such microphysics is beyond the scope of this paper, there should some discussion on the potential effects of the non-linearity of CCN numbers and emissions. In other words, the estimation of CCN based on peak emission in 1985 is likely to be significantly overestimated because higher condensation/coagulation sinks would prevent the growth of sub-CCN particles into CCN size range.*

Author Response: The fact, that an increase in emissions does not cause an increase of the concentrations of particle mass by the very same factor, is considered as well in our study. The increase of the SO_2 emission rates of a factor of 19 resulted in estimated mass concentration increases of 3.9 and 5.3 for ammonium sulfate and sulfuric acid, respectively, which in the end, under the assumption that the size distribution of each chemical species is the same in 1985 and 2013, results in roughly a factor of 2 increase in CCN number concentration. Although the size distributions are assumed to be the same in 1985 and 2013, the aerosol composition is different, and hence at least this non-linear effect of changed emissions on the calculated number of CCN is considered.

We are aware that the size distribution between the two scenarios 1985 and 2013 is likely different. The CCN concentrations might be overestimated for the 1985 scenario and should therefore be considered as an upper border estimate for the highly polluted period in Europe in the 1980s. In the revised manuscript we will discuss this more clearly.

Change in manuscript (p.8, l.14 - 18): Due to lack of observational data of aerosol size distributions in the 1980s in the study region to able to generalize 2010s and 1980s size distributions, for this study the same size distributions for 1985 and 2013 were assumed. Since the size distribution is crucial in order to translate modeled aerosol mass into number and finally derive

CCN number, this assumption is likely an important source of uncertainty, which is difficult to reliably quantify.

Referee Comment: *2. This study assumes ammonium nitrate concentration was zero in 1985. P6. L6 makes the following justification: "Particulate ammonium sulfate can be formed in the atmosphere from emitted sulfur dioxide and ammonia. In case there is still ammonia left after this reaction, ammonium nitrate can be formed as well. As can be seen from Tab. 1, almost 20 times more SO2 was emitted during the 1980s compared to 2013. For this reason, there was much more sulfate available in the atmosphere than necessary for the transformation of the total available ammonia to ammonium sulfate. This is why in this study the production of ammonium nitrate was set to zero and half of the additional sulfur was transformed to sulfuric acid for the 1985 scenario." Is there any literature to support this assumption? Why would not ammonium nitrate formation occur simultaneously with ammonium sulfate formation? According to their logic, when SO2 emission is very high, one would not observe nitrates in particles, but nitrates are commonly observed in a present-day polluted city with high SO2 with AMS measurements (e.g., Beijing) (Jimenez et al. 2009).*

Author Response: As mentioned by you, ammonium nitrate can also from under very high concentrations of sulfur. We oriented our calculations at the representation of the ammonium sulfate and ammonium nitrate formation in the model (Hinneburg et al., 2009). The implementation follows Simpson et al. (2003) who state (p. 39): "In the model ammonium sulphate is formed instantaneously from NH_3 and SO_4, only limited by the availability of the least abundant of the two species. Any excess NH_3 may then react with HNO_3, forming ammonium nitrate (NH_4NO_3) in an equilibrium reaction." Therefore the model would not form any ammonium nitrate in the 1985 scenario (see calculation in the response to the next comment). Ammonium nitrate is the only particulate nitrate compound formed in the model. Therefore, the model is likely to underestimate the nitrate concentration under conditions with high SO_2 emissions.
Furthermore, the density, size distribution and activation potential of ammonium sulfate and ammonium nitrate are similar. Therefore, exchanging part of the ammonium sulfate with ammonium nitrate would introduce only minor changes to the calculated CCN number concentration.
The revised manuscript will be improved with a more detailed explanation of the assumptions and calculation of the 1985 aerosol conditions.

Hinneburg, D., E. Renner, and R. Wolke (2009), Formation of secondary inorganic aerosols by power plant emissions exhausted through cooling towers in Saxony, Environ Sci Pollut R, 16(1), 25-35.

Simpson, D., et al. (2003), Transboundary acidification, eutrophication and ground level ozone in Europe PART I: Unified EMEP model description., EMEP Report 1/2003, Norwegian Meteorological Institute, Oslo, Norway.

Change in manuscript (p.6, l.20 - p.8, l.21): The whole section was revised.

Referee Comment: *3. This study assumes additional sulfur was transformed to sulfuric acid for the 1985 scenario (p7. L3). How does that lead to "Sulfate" value of AS_2013 * 5.3 in Table 3?*

Author Response: Since no access sulfuric acid after formation of ammonium sulfate was left in the 2013 scenario, scaling up the 2013 sulfuric acid mass could not be done. Therefore, assumptions were made to calculate the sulfuric acid mass for 1985 from the ammonium sulfate concentrations in 2013:

- In the model, the formation of ammonium sulfate is described by paring 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.
- In the 1985 scenario, no ammonia is left for the formation of ammonium nitrate and we assume that no ammonium nitrate is formed. As mentioned above, due to the similar

hygroscopic properties for ammonium sulfate and ammonium nitrate, this simplification is not expected to have significant impacts on the CCN concentration.

- 50% of the SO_2 leftover after ammonium sulfate formation is assumed to form sulfuric acid. This is a source of uncertainty and will be discussed in the revised manuscript.

The calculation of the scaling factors between 2013 and 1985 is as follows:

- 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 leftover SO_2 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.

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

Change in manuscript (p.6, l.20 - p.8, l.21): The whole section was revised.

Referee Comment: *4. Does "Sulfate" in Table 1 and 3 means "Sulfuric acid"?*

Author Response: Sulfuric acid is formed in the model. Due to its low vapor pressure, it is entirely partitioned to the particle phase as sulfate. The accounted molar mass is that of sulfate. We will make this clearer in the revised version to avoid confusion.

Change in manuscript (p.8, l.5 - 6): This is why in this study the production of ammonium nitrate was set to zero and half of the additional sulfur was transformed to sulfuric acid for the 1985 scenario. Sulfuric acid is assumed to entirely partition to the particulate phase and is therefore accounted for as sulfate.

Referee Comment: *5. Table 1: Shouldn't sigma (m) in the log-normal distribution be dimensionless?*

Author Response: Yes, this is correct and will be changed accordingly in the revised manuscript.

Change in manuscript: Changed accordingly in Table 1.

Referee Comment: *6. P8. Line 21. How are multiple-charge particles accounted for in in-situ CCNC measurements?*

Author Response: The monodisperse CCNc measurements were corrected for multiple-charged particles up to three charges. For the whole diameter size range the apparent connected diameter of doubly and triply charged particles was calculated via the linear relationship between single and doubly as well as between single and triply charged particles (Wiedensohler, 1988). For a given activation curve (CCN/CN) the fraction of apparently doubly and triply charged particles is than subtracted from the activated fraction. The resulting curve is valid for single charged particles and was fitted with a sigmoidal fit to get the activation diameter (50% of particles activated).

Change in manuscript (p.10, l.6-9): 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 particles 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: *7. Is there any additional information on the mini CCN counter other than "custom built by Gregory C. Roberts"? Has it been used in any prior published study?*

Author Response: This mini CCN instrument is working after the same principal 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).

Change in manuscript (p.10, l.15-18): 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).

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.

Referee Comment: *8. Conclusion: "In conclusion, the simulated profiles of the present-day simulation are within the variability range of the measurement-based profiles and thus represent realistic condition" This seems to be an over-statement. Figure 6 clearly shows the model and observation are outside 25-75% regions.*

Author Response: Thank you for the important comment. The section is re-written in the revised manuscript and will focus on the representation in the boundary layer. In the boundary layer model and observations agree mostly within a factor of two. However, the model generally overestimates the observations. Considering the factor of 2-3 uncertainty of the observed CCN number concentrations based on the LIDAR retrieval (Mamouri and Ansmann, 2016), we think that there is reasonable agreement within the boundary layer. For the few days of the in-situ CCN-profile observed by the helicopter-borne platform ACTOS, the model and observation 25-75% range do not overlap, however, the overestimation of a factor of 2 in the boundary layer is generally acceptable for the scope of the study.
In the free troposphere, the model more strongly overestimates the observations. In particular the sharp decrease in CCN number concentration at around 1.5 km height is not represented as sharp in the model. We will include a more detailed discussion in the revised manuscript.

Change in manuscript (p.20, l.7-9): Within the boundary layer, the simulated vertical profiles of the present-day simulation are within the variability range of the CCN derived from lidar measurements but do deviate from the in-situ helicopter-borne CCN measurements outside their 0.25-0.75 quantile range.

Referee Comment:
Typo
p.7 L22. Unnecessary comma (Spindler et al., 2013),.
p.12 L4 repeated "in"
p.14 Line 4. "cf. Figs.6 and ??" What is "??". Should it be 7?

Author Response: Thank you for these findings. The mentioned typos will be corrected in the revised manuscript.

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:

<u>*The calculation of the 1985 estimates*</u>

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.

Change in manuscript (p6, l20 - p8, l21): The whole section was revised.

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 individual 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 modeled total aerosol size distribution is combined of the ones of the individual species (which have different shapes and geometric mean radii) and is therefore multi-modal, and partly accounting for the Aitken mode range. The combined size distribution of the different species was found to match the observed size distribution well between 50 and 200 nm (i.e., the peak region of the observed accumulation mode), which is most relevant for estimating CCN. Applying a size-resolved aerosol transport model for a similar study would be a useful next step, which would also avoid uncertainty by using the same size distributions in the whole domain and for the 1980s and present day scenarios.

Change in manuscript (p.4, l.29 - p.5, l.13): The whole paragraph whose revised.

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 (p.10, l.6-9): 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 particles 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.

Change in manuscript (p.5, l.14 - p.6, l.9): The whole paragraph whose revised.

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 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.

Change in manuscript (p.6, l.10-14): The same method to derive CCN concentrations from the modeled aerosol mass as applied in this study was utilized in a related study of the HD(CP)^2 project to parameterize the CCN concentrations as a function of vertical velocity (Hande et al., 2016). As written above, they evaluated the aerosol size distribution at Melpitz and found good agreement in the CCN size range. We therefore assume that the applied method generally produces realistic CCN concentrations. However, the ambient aerosol size distribution varies in time and space and therefore the assumption of a spatially and temporally constant size distribution for the different aerosol species is a source of uncertainty.

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.

P11, 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.

Change in manuscript (p.12, l.11 - p.13, l.7): The whole paragraph was revised taking into account also other comments.

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).

Change in manuscript (p.14, l.3 - p.16, l.2): The whole section was revised taking into account also other comments.

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.

Change in manuscript (p.14, l.3 - p.16, l.2): The whole section was revised taking into account also other comments.

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.

Change in manuscript (p16, l.5 - p.17, l.16): The whole section was revised taking into account also other comments.

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.

Change in manuscript (p16, l.5 - p.17, l.16): The whole section was revised taking into account also other comments.

Referee Comment:

<u>4: References</u>

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.

Change in manuscript (p.2, l.33 - p.3, l.2 and bibliography): Such data sets can be used to evaluate the application of available aerosol activation parameterizations in atmospheric models. Evaluated against in-situ observations, the applied regional and global models (e.g., Spracklen et al., 2011; Bègue et al., 2015; Schmale et al., 2019; Fanourgakis et al., 2019; Watson-Parris et al., 2019) tend to underestimate the observed CCN concentrations.

Bègue, N., Tulet, P., Pelon, J., Aouizerats, B., Berger, A., and Schwarzenboeck, A.: Aerosol processing and CCN formation of an intense Saharan dust plume during the EUCAARI 2008 campaign, Atmospheric Chemistry and Physics, 15, 3497–3516, https://doi.org/10.5194/acp-15-3497-2015, https://www.atmos-chem-phys.net/15/3497/2015/, 2015.

Fanourgakis, G. S., Kanakidou, M., Nenes, A., Bauer, S. E., Bergman, T., Carslaw, K. S., Grini, A., Hamilton, D. S., Johnson, J. S., Karydis, V. A., Kirkevåg, A., Kodros, J. K., Lohmann, U., Luo, G., Makkonen, R., Matsui, H., Neubauer, D., Pierce, J. R., Schmale, J., Stier, P., Tsigaridis, K., van Noije, T.,Wang, H.,Watson-Parris, D.,Westervelt, D. M., Yang, Y., Yoshioka, M., Daskalakis, N., Decesari, S., Gysel-Beer,M., Kalivitis, N., Liu, X.,Mahowald, N.M., Myriokefalitakis, S., Schrödner, R., Sfakianaki,M., Tsimpidi, A. P.,Wu,M., and Yu, F.: Evaluation of global simulations of aerosol particle and cloud condensation nuclei number, with implications for cloud droplet formation, Atmospheric Chemistry and Physics, 19, 8591–8617, https://doi.org/10.5194/acp-19-8591-2019, https://www.atmos-chem-phys.net/19/8591/2019/, 2019.

Schmale, J., Baccarini, A., Thurnherr, I., Henning, S., Efraim, A., Regayre, L., Bolas, C., Hartmann,M.,Welti, A., Lehtipalo, K., Aemisegger, F., Tatzelt, C., Landwehr, S., Modini, R. L., Tummon, F., Johnson, J. S., Harris, N., Schnaiter,M., Toffoli, A., Derkani, M., Bukowiecki, N., Stratmann, F., Dommen, J., Baltensperger, U., Wernli, H., Rosenfeld, D., Gysel-Beer, M., and Carslaw, K. S.: Overview of the Antarctic Circumnavigation Expedition: Study of Preindustrial-like Aerosols and Their Climate Effects (ACE-SPACE), Bulletin of the American Meteorological Society, 100, 2260–2283, https://doi.org/10.1175/BAMS-D-18-0187.1, https://doi.org/10.1175/BAMS-D-18-0187.1,25 2019.

Spracklen, D. V., Carslaw, K. S., Pöschl, U., Rap, A., and Forster, P. M.: Global cloud condensation nuclei influenced by carbonaceous combustion aerosol, Atmospheric Chemistry and Physics, 11, 9067–9087, https://doi.org/10.5194/acp-11-9067-2011, https://www.atmos-chem-phys.net/11/9067/2011/, 2011.

Watson-Parris, D., Schutgens, N., Reddington, C., Pringle, K. J., Liu, D., Allan, J. D., Coe, H., Carslaw, K. S., and Stier, P.: In situ constraints on the vertical distribution of global aerosol, Atmospheric Chemistry and Physics, 19, 11 765–11 790, https://doi.org/10.5194/acp-19-11765-2019, https://www.atmos-chem-phys.net/19/11765/2019/, 2019.

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.

Change in manuscript (p.3, l.33 - p.4, l.2 and bibliography): COSMO is driven by initial and boundary data from GME re-analysis (the global model of DWD operational in 2013, Majewski et al., 2002).

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).

Change in manuscript (p.9, l.10-12 and bibliography): Added references.

Referee Comment: *P8 L34: 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.

Change in manuscript (p.3, l.9-10): Corrected brackets at first mentioning of HOPE.

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.

Change in manuscript (p.9, l.20-23 and bibliography): Added references.
Additionally, during the fall campaign, in-situ observations with the helicopter-borne platform ACTOS (Airborne Cloud Turbulence Observation System, Siebert et al., 2006) were combined cloud properties observed with remote sensing at the LACROS (Leipzig Aerosol and Cloud Remote Observations System, Bühl et al., 2013) supersite

Referee Comment: *P9, L12: 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 (p.10, l.15-18): 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.

Change in manuscript: Updated bibliography entry.

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 (p.12, l.11 - p.13, l.7): The whole paragraph was revised taking into account also other comments.

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

Author Response: On average the measured CCN concentrations are overestimated by 37 % (20.1 % without outliers) for the fall period, and underestimated by 29 % for the spring period by the model. We will add this information in the revised manuscript.

Change in manuscript (p.12, l.11 - p.13, l.7): The whole paragraph was revised taking into account also other comments.

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 (without the outliers) and 1.40, respectively. We will add this information to the plot and the text.

Change in manuscript (p.14, l.3 - p.16, l.2): The whole section was revised taking into account also other comments. Furthermore, regression lines have been added to Fig. 3 and the figure caption has been changed accordingly.

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.

Change in manuscript (p.14, l.3 - p.16, l.2): The whole section was revised taking into account also other comments.

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 up the factor of 2. 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.

Change in manuscript (p16, l.5 - p.17, l.16): The whole section was revised taking into account also other comments.

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.

Change in manuscript (p16, l.5 - p.17, l.16): The whole section was revised taking into account also other comments.

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.

Change in manuscript (p.14, l.3 - p.16, l.2): The whole section was revised taking into account also other comments. Table 4 now holds the average measured CCN number concentration, which previously was only given in the table caption.

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 up the factor of 2. In the revised manuscript we will re-write this section including quantitative measures of the deviation between model and observation.

Change in manuscript (p16, l.5 - p.17, l.16): The whole section was revised taking into account also other comments.

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.

Change in manuscript (p.14, l.3 - p.16, l.2): The whole section was revised taking into account also other comments.

Referee Comment: *P14, L12. 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

P10, 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) 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 (p.12, l.6-9): 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 (p.3, l.8-9): 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 (p.2, l.19-21): 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.

Change in manuscript (p.1, l.7 and p.3, l.3): This is now stated in the abstract and introduction.

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 (p.3, l.14-17): The resulting modeled CCN fields can be used in atmospheric models that do not treat aerosol transport explicitly 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.

Change in manuscript (p.2, l.2 - p.3, l.26): The introduction was revised giving the aim of the study and an overview of the methods.

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.

Change in manuscript (p.3, l.30 - p.4, l.11): The whole section was revised.

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.

Change in manuscript (p.4, l.24): In the vertical, the model treats 50 layers up to a height of 22km.

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, most of the data that was used was measured at Melpitz. From the observations at Jülich, only the derived CCN concentrations from lidar measurements were taken into account. In the revised manuscript we will change to using the terms “spring” and “fall period” instead of HOPE Jülich and HOPE Melpitz and include the Jülich site in Fig. 1.

Change in manuscript (Figure 1 and p.4, l.21-22): The caption of Fig. 1 was revised and the Jülich site is now shown in Fig. 1.

In addition, lidar-based CCN concentrations were available during the spring campaign in Jülich, Germany.

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.

Change in manuscript (p.4, l.15-17): In the ICON-LEM (ICOsahedral Non-hydrostatic Large Eddy Model; Zängl et al., 2015; Dipankar et al., 2015; Heinze et al., 2017), which is the model used in HD(CP)², there is no online aerosol transport scheme, which indicates the need of prescribing the aerosol and CCN concentrations in order to be considered for aerosol-cloud interaction.

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-LEM simulations within the framework of the HD(CP)² project. We will make this clearer in the revised manuscript.

Change in manuscript (p.4, l.18-20): In order to provide time varying 3D fields of CCN concentrations for ICON-LEM, model simulations with COSMO-MUSCAT covering most of Germany have been carried out for the time period of two intensive measurement campaigns during HD(CP)2: HOPE.

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.

Change in manuscript (p.5, l.1-3): For the externally mixed aerosols, the total number concentration of each species is calculated from the modeled mass of the aerosol species assuming an individual geometric mean radius and standard deviation.

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.

Change in manuscript (p.2, l.2 - p.3, l.26): The introduction was revised now giving information about why we choose the 1980s and the year 1985 in particular.

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. During the revision, we now found that on the original p.6, l. 7 the table reference was wrong. This is probably the position the Referee meant.

Changes in the manuscript (p.6, l.34): Corrected reference to Table 2.

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: Revised caption of table 4.

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 (CCN from ammonium nitrate: 1.0-1.5 x 10^9 m^{-3} for the gravimetrical measurements and 1.5-3.0 x 10^9 m^{-3} for the modelled CCN).

Change in manuscript (p.12, l.11 - p.13, l.7): The whole paragraph was revised taking into account also other comments.

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 displayed in Fig. 2.

Change in manuscript (p.12, l.11 - p.13, l.7): The whole paragraph was revised taking into account also other comments.

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.

Change in the manuscript (Fig. 6 and p.16, l.5-8): Updated Figure 6 and changed text:

Fig. 6 compares the simulated and observed vertical profiles of the CCN number concentration over Melpitz for the two periods in 2013. Fig. 6a shows the comparison to CCN derived from lidar observation during the spring period at Jülich, and Fig. 6b the comparison to the in-situ observations by the helicopter-based platform ACTOS during the fall period at Melpitz.

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).

Changes in the manuscript (p.18, l.4-5): For the calculation, a vertical velocity of 1 m s^{-1} was assumed. Hence, in contrast to the previous analysis the supersaturation depends on the aerosol composition and varies spatially and temporally.

And revised figure caption of Fig. 7.

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.

Estimation of Cloud Condensation Nuclei number concentrations and comparison to in-situ and lidar observations during the HOPE experiments

Christa Genz[1,a,*], Roland Schrödner[1], Bernd Heinold[1], Silvia Henning[1], Holger Baars[1], Gerald Spindler[1], and Ina Tegen[1]

[1]Leibniz Institute for Tropospheric Research (TROPOS), Permoserstraße 15, 04318 Leipzig, Germany
[a]now at German Centre for Integrative Biodiversity Research (iDiv) Halle-Jena-Leipzig, Deutscher Platz 5e, 04317 Leipzig, Germany
[*]previously published under the name Christa Engler

Correspondence: christa.genz@idiv.de

Abstract. Atmospheric aerosols are the precondition for the formation of cloud droplets and ~~have thus~~ therefore have large influence on the microphysical and radiative properties of clouds. In this work four different methods to derive ~~potential~~ or measure number concentrations of cloud condensation nuclei (CCN) ~~number concentrations~~ were analyzed and compared: (i) A model parameterization based on simulated particle concentrations, (ii) the same parameterization based on gravimetrical particle measurements, (iii) direct CCN measurements with a CCN counter ~~at a certain observation site and lidar derived~~ and (iv) lidar-derived and in-situ measured vertical CCN profiles. In order to allow for sensitivity studies of the anthropogenic impact, a scenario for the maximum CCN concentration under peak aerosol conditions (exemplary for the year 1985) was estimated as well. In general, the simulations are in good agreement with the observation. At ground level, an average value of around ~~$1*10^9$~~ 1×10^9 $\mathrm{CCN\,m^{-3}}$ at a supersaturation of 0.2 % was found with all methods. The discrimination of the chemical species revealed an almost equal contribution of ammonium sulfate and ammonium nitrate to the total number of ~~potential~~ CCN. This was not the case for the peak aerosol scenario, ~~where almost no nitrate particles were formed . The potential activation~~ in which it was assumed that no ammonium nitrate was formed while large amounts of sulfate present consuming all ammonia during ammonium sulfate formation. The CCN number concentration at five different supersaturation values has been compared to the measurements. The discrepancies between model and in-situ observations were lowest for the lowest and highest supersaturations, since chemical composition and the size distribution of the particles are less important in ~~this range~~these ranges. In the ~~mid supersaturation~~ mid-supersaturation regime, the model overestimated the potentially activated particle fraction by around 30 ~~%.~~ %. By comparing the simulation with observed profiles, the vertical distribution of the CCN concentration was found to be overestimated by up to a factor of 2 in the boundary layer. The analysis of the modern (year 2013) and the peak aerosol scenario (year 1985) resulted in a scaling factor, which was defined as the quotient of the average vertical profile of the peak aerosol and present day CCN concentration. This factor was found to be around 2 close to the ground, increasing to around 3.5 between 2 and 5 km and approaching 1 (i.e., no difference between present day and peak

aerosol conditions) with further increasing height. ~~By comparing the simulation with observed profiles, the vertical distribution of the potential CCN was found to be reasonable.~~

1 Introduction

Compared to today's atmosphere, in the 1980s the aerosol load was much higher (Smith et al., 2011; Cherian et al., 2014). Presumably, during this time the anthropogenic emissions of aerosols and precursor gases in Central Europe were at their maximum. At least since the 1990s aerosol concentrations in Central Europe have been decreasing.

Atmospheric aerosol particles play an important role in the microphysical processes of cloud formation and thus have a potentially large influence on cloud properties. However, the evaluation of their effects shows still large uncertainties (e.g., Boucher et al., 2013). In order to reduce those uncertainties, parameterizations to estimate the number concentrations of the ~~cloud condensation nuclei (CCN)~~ CCN have been developed for application in models. For a realistic simulation of ~~cloud adjustment and~~ microphysical aerosol-cloud-interactions and macroscopic cloud adjustment due to aerosol perturbations, a detailed representation of the aerosol in the models is required. To describe the activation of aerosol particles, the chemical composition, the number concentration and the size distribution of the aerosol particles have to be known. Parameterizations of the cloud droplet activation (e.g., Abdul-Razzak et al., 1998; Abdul-Razzak and Ghan, 2000; Petters and Kreidenweis, 2007) ~~utilize~~ apply the Köhler-Theory (Köhler, 1936) and have been implemented into regional chemistry transport models ~~(CTM; e.g., Bangert et al., 2011; Hande et al., 2016)~~(e.g., Bangert et al., 2011; Hande et al., 2016). The influence of the ~~aerosol composition on the droplet activation~~ droplet activation on the aerosol composition is described using the aerosol hygroscopicity, e.g., represented by the hygroscopicity parameter kappa (κ). These parameterizations enable the investigation of the interaction of the aerosol population with cloud microphysical properties.

For the regional ~~Chemistry Transport Model~~ chemistry transport model (CTM) that is used in this study ~~(COSMO-MUSCAT, see section 2.1.1),~~ (COSMO-MUSCAT, Wolke et al., 2012, see section 2.1.1), Sudhakar et al. (2017) extended the model system to allow aerosol-cloud-interactions applying the two-moment cloud microphysics scheme by Seifert and Beheng (2006). ~~Here,~~ This model version is online interactively coupled, making the activation of aerosol ~~particles to cloud droplets is directly considered by an online coupling of meteorology and aerosol transport~~mass available for the two-moment scheme. However, the aerosol ~~is treated as~~ activation uses the bulk mass and does not explicitly consider online computed aerosol microphysical properties. The complex consideration of aerosols and aerosol-cloud-interactions including the particle size distribution and composition in models is expensive with regard to ~~computating~~ computation time and storage and thus not feasible in particular for long-term applications.

Therefore, Hande et al. (2016) applied a combination of two existing models to produce a CCN climatology for use in ~~limited-aera~~ limited-area models, representing normal background conditions over Europe. First, the aerosol particle mass concentrations were simulated using a CTM with a mass-based aerosol scheme~~, and then~~. Then, the particle size distribution and the ~~potential activation of the particles to cloud droplets~~ CCN number concentration were calculated offline using the parametrization of Abdul-Razzak and Ghan (2000). For applying this activation parametrization ~~, the aerosol is assumed to be~~

~~externally mixed, and the number~~ on the modeled aerosol mass, the number distribution, size and chemical composition of the aerosol particles have to be prescribed.

Measurements of the CCN number concentration in the field are valuable in order to evaluate the ability of the models to describe the activation of aerosol particles. There are several studies of in-situ observations (e.g., Henning et al., 2014; Hammer et al., 2014; Friedman et al., 2013). Also the derivation of vertical profiles of CCN with ground-based remote sensing methods is possible ~~(e.g., Mamouri and Ansmann, 2016; Ghan et al., 2006). Direct comparisons of model results and measurements, however, are sparse~~(e.g., Ghan et al., 2006; Mamouri and Ansmann, 2016). Such data sets can be used to evaluate the application of available aerosol activation parameterizations in atmospheric models. Evaluated against in-situ observations, the applied regional and global models (e.g., Spracklen et al., 2011; Bègue et al., 2015; Schmale et al., 2019; Fanourgakis et a to underestimate the observed CCN concentrations.

~~In this study , a similar approach as in Hande et al. (2016) was applied to derive CCN from modeled aerosol distributions. It is part~~ The aim of this study is to provide estimates of the concentrations of cloud condensation nuclei (CCN) in the 1980s (with the year 1985 as a reference) over Germany and compare those to simulations and observations in the year 2013. The derived time varying 3D-CCN fields were used as input for high-resolution simulations over Germany in the framework of the High Definition Clouds and Precipitation for advancing Climate Prediction (HD(CP)^2) project ~~.~~ (see Heinze et al., 2017; Costa-Surós et al., 2019 A similar approach as in Hande et al. (2016) was applied to derive CCN from modeled aerosol mass concentrations. The aerosol particle concentrations were simulated using the regional CTM COSMO-MUSCAT with a mass-based aerosol scheme ~~(Wolke et al., 2012) for the~~ for two periods of the ~~(~~HD(CP)^2 Observational Prototype Experiments ~~(HOPE), Macke et al. (2017)) in 2013 and the peak aerosol scenario for the year 1985. Afterwards,~~ (HOPE, Macke et al., 2017) in 2013. Based on the modeled aerosol mass concentrations, the particle number size distributions ~~and potential activation~~ for each aerosol species and CCN number concentrations were calculated offline using the activation parametrization by Abdul-Razzak and Ghan (2000). ~~For the potential~~ The activation of aerosol particles ~~,~~ depends on their number, size, and chemical composition ~~were taken into account~~as well as the applied supersaturation (either fixed or derived from updraft velocities). Thus, this approach is very versatile and can be applied for each type of aerosol mixture. ~~We aim for a detailed description of the CCN abundance with three different methods: (i) model-derived, (ii) in-situ measurements, and (iii) ground-based remote sensing. The resulting~~ The resulting modeled CCN fields can be used in atmospheric models that do not treat aerosol transport explicitly to analyze clouds and their radiation effects~~with~~ . For this purpose, CCN fields of a variable degree of complexity ~~. This implies a fixed CCN profile~~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 ~~temporary~~ temporally limited episodes. ~~In detail, the following CCN datasets were comparedin this study~~For the year 2013, the CCN number concentrations derived or measured with four different methods were compared: (i) ~~4D parameterization based on simulated present day (year 2013) particle~~ CCN derived from COSMO-MUSCAT simulations of aerosol mass concentrations, (ii) ~~4D parameterization based on simulated peak emission scenario (year 1985) particle mass concentrations~~CCN derived from gravimetrical aerosol mass measurements, (iii) ~~parameterization based on the measured particle mass concentrations,~~ ground-based in-situ measurements of CNC, and (iv) ~~CCN concentrations measured directly with~~

~~the CCN counter, and (v)~~ vertical CCN profiles ~~obtained from~~ derived from ground-based lidar remote sensing and observed by helicopter-borne in-situ measurements.

In order to estimate the CCN concentrations in the example year 1985, the aerosol concentrations from the 2013 simulation were scaled based on 1980s emission estimates. The derived CCN fields for 1985 were compared to the 2013 simulation and the observations of the year 2013.

The manuscript is structured as follows. First, the applied CTM COSMO-MUSCAT as well as the different observation techniques are introduced and necessary assumptions are described. In section 3, the results of the comparison of CCN number concentrations obtained from the different methods are discussed. Conclusions and a summary can be found in section 4.

2 Methods

2.1 Model description

2.1.1 COSMO-MUSCAT

For this study, the chemistry transport model system COSMO-MUSCAT (Wolke et al., 2012) was used. It consists of the meteorological model COSMO (COnsortium for Small scale MOdelling), which is the operational forecast model of the German Weather Service (DWD), and the chemistry transport model MUSCAT (MUltiScale Chemistry Aerosol Transport). ~~MUSCAT is coupled to COSMO after a 24h spin-up time in each composition cycle. COSMO~~ COSMO is driven by initial and boundary data from GME re-analysis ~~. To ensure a realistic description of the~~ (the global model of DWD operational in 2013, Majewski et al., 2002). After a spin-up phase for COSMO of 24 hours, both models run coupled online for 48 hours. To ensure to stay close to the real meteorological conditions, ~~COSMO was reinitialized every 48 hours. Both models are coupled online ; the meteorology from COSMO drives~~ the model system is then re-initialized for the next simulation cycle. The online coupling has the advantage that the meteorological fields from COSMO are forwarded to MUSCAT in every time step. The meteorological fields drive the chemical transformation and atmospheric transport ~~,~~ treated in MUSCAT for several gas phase ~~species and aerosol particle populations~~and aerosol species. Transport processes include advection, turbulent diffusion, sedimentation, dry and wet deposition. MUSCAT is based on mass balances, which are described by a system of time-dependent, three-dimensional advection-diffusion reaction equations. Emissions of anthropogenic primary particles and precursors of secondary aerosols are prescribed using emission fields from EMEP ~~(European Monitoring and Evaluation Programme; EMEP (2009))~~(European Monitoring and Evaluation
Emissions of natural primary aerosols (Saharan desert dust, primary marine aerosol particles) are computed within the model (e.g., Heinold et al., 2011), using meteorological fields (surface wind speed, precipitation) from the model itself in addition to information on surface properties~~read from satellite products~~.

2.2 Model setup

The study presented here is part of the High Definition Clouds and Precipitation for advancing Climate Prediction (HD(CP)^2) project. The main objective is to improve our understanding of clouds and precipitation, using a model for very high resolution simulations. In the ~~ICON-LES~~ICON-LEM (ICOsahedral Non-hydrostatic Large Eddy Model; Zängl et al., 2015; Dipankar et al., 2015; He which is the model used in HD(CP)^2~~(Dipankar et al., 2015; Heinze et al., 2017)~~, there is no online aerosol transport scheme, which indicates the need of ~~describing~~ prescribing the aerosol and CCN ~~offline,~~ concentrations in order to be considered for aerosol-cloud interaction.

~~Model simulations~~ In order to provide time varying 3D fields of CCN concentrations for ICON-LEM, model simulations with COSMO-MUSCAT covering most of Germany have been carried out for the time period of two intensive measurement campaigns during HD(CP)^2: HOPE~~, which were performed in Jülich and Melpitz, Germany~~. These campaigns cover the time periods between April 3 to May 31 ~~(Jülich)~~ and September 1-~~–~~30, 2013 (~~Melpitz,~~ see section 2.3). Data from the measurement site Melpitz, Germany, were used for comparison during both campaigns. In addition, lidar-based CCN concentrations were available during the spring campaign in Jülich, Germany.

The model domain investigated in this study is displayed in Fig. 1 and covers the area between 6-15°E and 48.25-54°N. The horizontal resolution was set to 7 km~~and the temporal resolution for the model output was set to 1h. 32~~. In the vertical, the model treats 50 layers up to a height of ~~8~~22 km~~were considered in the analysis~~. The temporal resolution for the model output was set to 1h. Besides the standard meteorological model output from COSMO, MUSCAT provides the mass concentrations of several gas phase ~~species and particulate compounds~~and aerosol species.

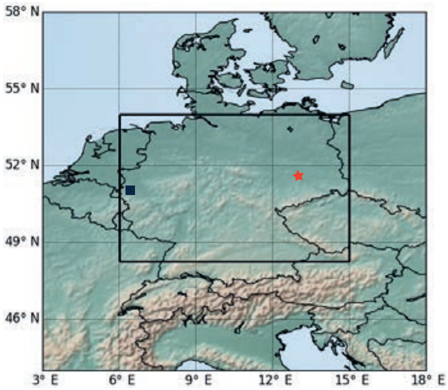

Figure 1. Model domain over Germany, which was used in this study. The red star marks the research station Melpitz (12.93°E, 51.53°N) and the blue square the measurement site Jülich (50.88°N, ~~86 m a.s.l.~~6.41°E).

2.2.1 Aerosol particle number estimation and CCN parametrization

Using the aerosol bulk scheme of COSMO-MUSCAT, the mass concentrations for the species considered are simulated. In order to compare the model results with in-situ particle measurements and to calculate number concentrations of ~~potential~~ CCN, particle number size distributions (PNSD) have to be estimated from those mass concentrations. For each species of the anthropogenic aerosol (ammonium sulfate (AS), ammonium nitrate (AN), sulfate (SU), organic (OC) and elemental carbon (EC)) and sea salt (SS), individual log-normal size distributions are assumed. The size distribution of the mineral dust (DU) particles follows a sectional scheme (Heinold et al., 2011). A log-normal size distribution is explicitly defined with the three parameters diameter or radius (d or r, respectively), standard deviation (σ) and total number concentration (N). ~~The~~ For the externally mixed aerosols, the total number concentration ~~can be~~ of each species is calculated from the ~~particle mass assuming spherical particles of a certain size and density. Then,~~ modeled mass of the aerosol species assuming an individual geometric mean ~~diameter~~ radius and standard deviation~~was assumed for each considered species~~. The choice of ~~geometric mean diameter and standard deviation defines the size distribution~~these parameters defines the aerosol number size distribution and is a critical source for uncertainty of aerosol and CCN number concentrations. Within the HD(CP)^2 framework, literature values, aerosol mass spectrometer (AMS) measurements~~from the TROPOS site Melpitz (Poulain et al., 2011), which is representative for central Europe (e.g., Spindler et al., 2012; Engler et al., 2007)~~ , and particle number size distribution measurements in the diameter range $10\,\mathrm{nm}$ to $10\,\mu\mathrm{m}$ from the TROPOS site Melpitz, Germany (Poulain et al., 2011), which is representative for central Europe (e.g., Spindler et al., 2012; Engler et al., 2007), were used to define the parameters for the log-normal ~~distribution estimation~~distributions. Adding up the ~~log-normal~~ different size distributions of all considered species gives the total particle number size distributions. The calculations have been compared to observational data and showed a good agreement ~~(Hande et al., 2016)~~to the observed total size distribution at Melpitz between 50 and $200\,\mathrm{nm}$ (Hande et al., 2016), which is most relevant for estimating CCN. The geometric mean ~~diameter~~radius, standard deviation and density for characterizing the particle number size distributions of the individual aerosol species is listed in Tab. 1, mostly according to the values used in Hande et al. (2016).

The number ~~and mode information of the particles could now be~~ size distributions of the aerosol species was now used to calculate the number of activated particles under certain conditions. ~~For this purpose, the individual~~ The calculation of the CCN number concentration in this study follows the parameterization of Abdul-Razzak and Ghan (2000) for multi-modal aerosol distributions, which relates the particle number size ~~distributions for each considered component of the aerosol were parameterized using~~ distribution and composition to the number of activated particles as a function of supersaturation. The individual aerosols compete for the available liquid water, determining the maximum supersaturation, which apart from the aerosol composition and individual size distributions depends on the updraft velocity. Abdul-Razzak et al. (1998) describe the parameterization for a single log-normal mode of aerosol particles (only for a single species), whereas Abdul-Razzak and Ghan (2000) deve an extended approach for multiple soluble and insoluble aerosol species, representing a multi-modal aerosol size distribution. The parameterization uses the hygroscopicity parameter κ ~~individually~~of each considered aerosol species. The κ values used in this study can be found in Tab. 1 as well. κ was defined first in Petters and Kreidenweis (2007) as a single parameter to describe the relationship between the particle dry diameter, its hygroscopicity, and the CCN activation. In several laboratory studies, κ has been determined experimentally. Highly hygroscopic particles can have a $\kappa > 1$, while for totally hydrophobic particles $\kappa = 0$. Petters and Kreidenweis (2007) reported κ for a number of different compounds, e.g., ammonium sulfate being about 0.6 in the supersaturation regime. Further studies investigated κ for other substances like sea salt (e.g., Niedermeier et al., 2008), coated soot (e.g., Henning et al., 2010) and secondary organic aerosol (e.g., Wex et al., 2009; Duplissy et al., 2011) or ~~depending~~ in dependence on the mixing state of the particles (Wex et al., 2010).

The ~~calculation of the CCN number concentration~~ same method to derive CCN concentrations from the modeled aerosol mass as applied in this study ~~follows the parameterization of Abdul-Razzak and Ghan (2000), which relates the particle number size distribution and composition to~~ was utilized in a related study of the ~~number of activated particles~~ HD(CP)^2 project to parameterize the CCN concentrations as a function of ~~supersaturation. To achieve the maximum supersaturation (as a function of vertical velocity), accounting for particle growth before and after activation, the supersaturation balance is used. Abdul-Razzak et al. (1998) describe the parameterization for a single lognormal mode of aerosol particles (only one single species), whereas Abdul-Razzak and Ghan (2000) proposed an extended version for multiple soluble and insoluble material. Here, two (or more) particle modes compete for the available water. The same method was utilized in a previous study for parameterizing the CCN concentrations as a function of vertical velocity (Hande et al., 2016). The model is assumed to produce realistic supersaturation fields and thus also realistic CCN numbers for stratiform clouds. For convective clouds, this is different, since the sub-grid supersaturation can be much higher than the grid cell average (e.g., Hill et al., 2015).~~ vertical velocity (Hande et al., 2016). As written above, they evaluated the aerosol size distribution at Melpitz and found good agreement in the CCN size range. We therefore assume that the applied method generally produces realistic CCN concentrations. However, the ambient aerosol size distribution varies in time and space and therefore the assumption of a spatially and temporally constant size distribution for the different aerosol species is a source of uncertainty.

In order to evaluate ~~the estimations, the number concentrations of the CCN~~ these assumptions, the modeled CCN number concentrations were compared to measurements close to the ground for the TROPOS super-site Melpitz. For this purpose, the same supersaturations as applied in the CCN number concentration measurements with ~~the~~ a cloud condensation nucleus counter ~~(CCNC, Henning et al. (2014))~~ (CCNC, Henning et al., 2014) were applied to the simulated particle number size distributions (see section 2.3.1).

2.2.2 Estimation of peak aerosol in 1985

In order to allow for sensitivity studies ~~of the anthropogenic impact , a peak aerosol scenario for~~ on the impact of anthropogenic pollution on CCN concentrations, a scenario to estimate aerosol concentrations for the year 1985 was developed. Due to the maximum emissions of aerosols and precursor gases in Europe during the 1980s, the year 1985 was taken as a reference year to compare to modern conditions. ~~The annual emissions of sulfur dioxide and ammonia during the years 1985 and 2013 (see Tab. 2) served as basis for these estimations (Hausmann, 2017). In the beginning~~ In the early 1990s, ~~environment~~ environmental protection became much more important, so efficient emission reduction strategies were developed. Furthermore, many aerosol and precursor sources simply disappeared after the liquidation of several ~~industrie~~ industry sites in Eastern ~~Europe~~ Germany and the former East-bloc countries after the political change in 1990.

The ~~assumptions made~~ calculations for 1985 were carried out offline with the model run from 2013 as a basis. The annual emissions of sulfur dioxide and ammonia during the years 1985 and 2013 (see Tab. 2) were utilized for these estimations (Hausmann, 2017, Umweltbundesamt (UBA, German Federal Environmental Agency), personal communication). The scaling factors derived in order to estimate the aerosol concentrations in 1985 based on the present day simulation are summarized in Tab. 3. The model implementation of the formation of ammonium sulfate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3)

Table 1. Physical and chemical aerosol properties used in this study. The values for the particle radius and standard deviation of the size distribution follow Poulain et al. (2011)~~and~~ , Spindler et al. (2012) (non-dust species) and Heinold et al. (2011) (mineral dust). Several laboratory and model studies served as basis for the κ values used in this study (Ghan et al., 2001; Petters and Kreidenweis, 2007; Wex et al., 2009; Duplissy et al., 2011).

Species	κ	σ ~~()~~	r (µm)	ρ (kgm^{-3})
Ammonium sulfate	0.51	1.6	0.05	1.77
Ammonium nitrate	0.54	1.6	0.05	1.725
Sulfate	1	1.6	0.05	1.8
Sea salt 1	1.16	1.8	0.065	2.2
Sea salt 2	1.16	1.7	0.645	2.2
EC	~~5*10^{-7}~~ 5×10^{-7}	1.8	0.03	1.8
OC	0.14	1.8	0.055	1.0
Mineral dust 1	0.14	2.0	0.2	2.65
Mineral dust 2	0.14	2.0	0.6	2.65
Mineral dust 3	0.14	2.0	1.75	2.65
Mineral dust 4	0.14	2.0	5.25	2.65
Mineral dust 5	0.14	2.0	15.95	2.65

Table 2. Annual emissions of dust, sulfur dioxide and ammonia for entire Germany during 1985 and 2013 in Mt ~~(Hausmann, 2017)~~as provided by Umweltbundesamt (German Federal Environmental Agency, UBA, Hausmann, 2017, personal communcation). So called dust also includes e.g., soot and resuspended material besides the natural mineral dust. The table also includes the factors, which the concentrations in 2013 are scaled with in order to estimate the concentrations in 1985.

	1985	2013	ratio 1985/2013
dust (incl. soot)	2.65	0.35	7.7
SO_2	7.73	0.41	19
NH_3	0.86	0.74	1.2

is described by Hinneburg et al. (2009) and follows Simpson et al. (2003). Particulate ammonium sulfate can be formed in the atmosphere from ~~emitted sulfur dioxide~~ sulfuric acid (formed after oxidation of SO_2) and ammonia. In the model, first ammonium sulfate is formed until either ammonia or sulfuric acid is consumed. In case there is still ammonia left after this reaction, ammonium nitrate can be formed as well. As can be seen from Tab. ~~1~~2, almost 20 times more SO_2 was emitted in Germany during the 1980s compared to ~~2013.~~ 2013, whereas NH_3 emissions remained almost unchanged. For this reason, there was much more ~~sulfate~~ sulfuric acid available in the atmosphere than necessary for the transformation of the total available

Table 3. Assumptions for the estimation of the aerosol conditions for the 1980s over Germany.

	2013	1985
Ammonium sulfate	AS_{2013}	AS~~$_{2013}$*3.9~~ $_{2013} \cdot 3.9$
Ammonium nitrate	AN_{2013}	0
Sulfate	SU_{2013}	AS~~$_{2013}$*5.3~~ $_{2013} \cdot 5.3$
EC	EC_{2013}	EC~~$_{2013}$*2~~ $_{2013} \cdot 2$
OC	OC_{2013}	OC_{2013}
Sea salt	SS_{2013}	SS_{2013}
Mineral dust	DU_{2013}	DU_{2013}

ammonia to ammonium sulfate. ~~This is why in this study~~ In 2013, SO_2 and NH_3 react to ammonium sulfate until SO_2 is consumed leading to the formation of 0.85 Mt ammonium sulfate. In 1985, in the implemented scheme, first NH_3 is consumed and in total 3.32 Mt ammonium sulfate are formed. This results in a scaling factor for ammonium sulfate of 3.9. In this SO_2 limited regime in 2013, there would not be any NH_3 left to produce ammonium nitrate. The inhomogeneous distribution and the time-dependent formation would still enable nitrate formation in reality. However, since assumed density, size distribution and hygroscopicity of ammonium sulfate and ammonium nitrate are similar, exchanging part of the ammonium sulfate with ammonium nitrate and vice versa would not introduce strong changes to the calculated CCN number concentration, which is the aim of this study. This is why, the production of ammonium nitrate was set to zero ~~and half of the additional sulfur was transformed to sulfuric acid~~ for the 1985 ~~scenario. This approach is~~ scenario. The ammonium sulfate formation leaves 6.1 Mt SO_2 unconsumed. Half of this excess SO_2 left after ammonium sulfate formation in 1985 is assumed to be oxidized to sulfuric acid. Sulfuric acid is assumed to entirely partition to the particulate phase and is therefore accounted for as sulfate. The approach described above is also encouraged by the serious ~~"acid rain "~~ acid rain problem in the 1980s (e.g., Seinfeld and Pandis, 1998, p. 1030ff). Since no excess sulfate is present in the 2013 simulation, we calculate the 1985 sulfate concentration based on the 2013 ammonium sulfate concentration. The ratio between the formed sulfate in 1985 (4.68 Mt) and the formed ammonium sulfate in 2013 (0.85 Mt) results in a scaling factor of 5.3. Since no emission data for elemental carbon in 1985 were available, the particle concentrations were assumed to be twice as high as in 2013. This is only justified by the fact that aerosol concentrations in the 1980s over Central Europe were higher than today mainly caused by combustion processes for heating and energy production. Organic carbon, sea salt and dust are supposed to result mostly from natural sources and thus ~~set to the values from 2013 in~~ remain unchanged for the 1985 ~~as well.~~ scenario.

Due to lack of observational data of aerosol size distributions in the 1980s in the study region to generalize size distributions of the 2010s and 1980s, for this study the same size distributions for 1985 and 2013 were assumed. Since the size distribution is crucial in order to translate modeled aerosol mass into particle numbers and finally derive CCN numbers, this assumption is likely an important source of uncertainty, which is difficult to quantify reliably.

The ~~calculations for 1985 were carried out offline with the model run from 2013 as a basis, since there was no detailed emission data for 1985 available. This~~ above scaling approach implies, that the meteorological conditions of the year 1985 were not taken into account. The results have to be interpreted carefully and ~~only as~~ represent only a rough estimate for the 1980s, ~~not for~~ but may be not representative for the specific conditions in spring and fall ~~1985 in particular.~~ 1985. The results of the comparison of the number concentrations in 2013 and 1985 ~~, which represents the peak aerosol over Europe,~~ are presented in section 3.

2.3 Measurements during HOPE

The present study utilizes observational data from the extensive measurements conducted during ~~both HOPE campaigns~~ the two HOPE campaigns (April 3 to May 31 and September 1 - 30, 2013) at the TROPOS research station Melpitz ~~. Additionally, at this site~~ and the measurement site near Jülich, Germany. At Melpitz additonal long-term measurements of in-situ aerosol PNSD, CCN concentrations and chemical composition of the aerosol particles are available. The rural-background site Melpitz (12.93°E, 51.53°N, 86 m a.s.l.) is located in Germany, ∼~~30~~40 km east of Leipzig ~~(Spindler et al., 2013; Engler et al., 2007)~~ in the East German lowlands. The site at a meadow is surrounded by agricultural land. It is representative for a large area in Central Europe and long-term studies with consideration of marine or continental air mass inflow enables the investigation of the influence of different spatially distributed emission sources and long-range transport on particulate matter (PM) concentrations ~~(Spindler et al., 2013),~~(Engler et al., 2007; Spindler et al., 2013). The Melpitz site is integrated in the infrastructure network ACTRIS (Aerosols, Clouds, and Trace gases Research Infrastructure Network, www.actris.eu) and EMEP ~~(Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe)~~(Co-operative Programme for Mon
From the spring campaign at Jülich only the lidar measurements were used to derive vertical profiles of CCN concentrations.

The idea behind ~~HOPE~~ the HOPE campaigns was to gain a comprehensive dataset of observations for evaluation of the new German operational forecast model ICON ~~(ICOsahedral Nonhydrostatic)~~ at the scale of a couple hundred meters (ICON-LEM). The campaign focused on the convective atmospheric boundary layer, especially the connection of clouds and precipitation. Technically, HOPE aimed at combining most of the surface flux and mobile ground-based remote-sensing observations available in Germany within a single domain for the purpose of describing the vertical structure and horizontal variability of wind, temperature, humidity, aerosol particles and cloud droplets in a high temporal and spatial resolution.

~~In the HOPE-Melpitz~~ Additionally, during the fall campaign, in-situ observations with the helicopter-borne platform ACTOS ~~(Airborne Cloud Turbulence Observation System)~~ (Airborne Cloud Turbulence Observation System, Siebert et al., 2006) were combined with aerosol and cloud properties observed with remote sensing at the LACROS ~~(Leipzig Aerosol and Cloud Remote Observations System)~~ (Leipzig Aerosol and Cloud Remote Observations System, Bühl et al., 2013) supersite. This dataset allows for the investigation of the relationship between tropospheric clouds and aerosol conditions.

Detailed information on the meteorological conditions during the two campaigns can be found in ~~Macke et al. (2017)~~ Macke et al. (2017)
Tab. 3 and 4. The weather situations during ~~HOPE-Jülich~~ the spring campaign changed from a few high-pressure systems with high-level cirrus clouds, interrupted by several frontal passages (warm and cold fronts) at the beginning of the campaign, and

followed by more shallow convective clouds later on. ~~HOPE-Melpitz~~ The fall period was dominated by low-level overcast clouds.

2.3.1 In-situ CCNC measurements - ground-based and airborne

Ground-based in-situ measurements with the CCNC are operational in Melpitz since August 2012 (Schmale et al., 2017) and the results were available for model evaluations within this study. The ambient CCN number concentration at Melpitz station was determined by means of size segregated activation measurements as described in detail in Henning et al. (2014), following the ACTRIS SOP (standard operating procedures, Gysel and Stratmann, 2013). Briefly, the set-up is as follows, downstream of the aerosol inlet and the drier unit, an aerosol flow of $1.5\,\mathrm{L\,min^{-1}}$ is size-selected with a DMPS system (Differential Mobility Particle Sizing system) and afterwards divided between a condensation particle counter ($1\,\mathrm{L\,min^{-1}}$ working flow, CPC 3010, TSI Aachen Germany) and a cloud condensation nucleus counter ($0.5\,\mathrm{L\,min^{-1}}$ working flow, CCNC, CCN-100, Boulder, USA). With the CCNC, a stream-wise thermal gradient cloud condensation nucleus counter (Roberts and Nenes, 2005), the supersaturation-dependent activation of the particles is investigated at 0.1, 0.2, 0.3, 0.5, 0.7 and 1 % supersaturation. 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. ~~This~~ The AF was corrected for multiply charged particles 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. This curve is fitted with a sigmoidal function describing the activation curve with the four parameters ~~–~~lower activation limit, upper limit, sigma (σ) and the critical diameter (D_c). Multiplying the activation curve (CCN/CN) with the ambient size distribution integral results in the ambient CCN number concentration at the given supersaturation. One measurement per supersaturation is available every two hours.

During ~~HOPE-Melpitz~~ the fall measurement campaign of HOPE also the helicopter-borne measurement platform ACTOS was deployed ~~(Siebert et al., 2006)~~ in Melpitz. The experimental set-up and the flight characteristics are described in detail ~~in~~ by Düsing et al. (2018). 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).~~(mCCNc, custom built by Gregory C. Roberts, working principal as described b which has been applied successfully on ACTOS before (e.g., Wex et al., 2016). The miniCCNc measured the CCN number concentration at a supersaturation of 0.2 %. Vertical profile measurements are available for 8 flights between ~~Sept~~ September 12~~and Sept~~ -27, 2013.

2.3.2 Daily PM_{10} sampling at Melpitz site

Particles with aerodynamic diameter up to $10\,\mu\mathrm{m}$ (PM_{10}) were sampled daily at the Melpitz site. PM-High-Volume quartz filter samples for PM_{10} were collected using a High-Volume sampler (DIGITEL DHA-80, Walter Riemer Messtechnik, Germany), having a sampling flux of about $30\,\mathrm{m^3h^{-1}}$. The filter type is a MK 360 quartz fibre filter (Munktell, Grycksbo, Sweden). The measurement techniques ~~in order~~ to determine the particle mass, water soluble ions and carbonaceous particles are described by Spindler et al. (2013, 2012). The particle mass determination was performed gravimetrically. The conditioned filters (72 hours at 20°C and 50 % relative humidity) were weighted with a microbalance as tare (blank) and after sampling of particles as gross weight. Main water-soluble ions (NO^{3-}, SO_4^{2-}, Cl^-, Na^+, NH^{4+}, K^+, Mg^{2+}, Ca^{2+}) were analyzed by ion chromatography. The determination of organic and elemental carbon (OC and EC) was performed by a two-step thermographic method using a carbon analyzer (behr Labor-Technik, Germany). OC was vaporized at 650°C for 8 minutes under nitrogen atmosphere and catalytically converted to CO_2 and the remaining EC was combusted further in 8 minutes with O_2 at 650°C. The formed CO_2 was than quantitatively determined by a non-dispersive infrared detector (modified German standard VDI method 2465 part 2).

2.3.3 CCN concentrations derived by lidar measurements

During the HOPE campaigns, PollyXT lidar systems (Engelmann et al., 2016) were used to measure automatically and continuously the vertical state of the atmosphere in terms of aerosol particles and clouds. Lidar observations were performed in Melpitz ~~starting September 1 until 30 2013~~ (fall campaign) and Jülich (spring campaign) with the 12 channel-multiwavelength-polarization lidar PollyXT_OCEANET. Hourly averaged profiles of the particle backscatter and extinction coefficient as well as the particle depolarization ratio were calculated automatically for the whole measurement period as described in Baars et al. (2016). As the particle depolarization ratio was close to zero (indicator for spherical particles) for the whole period, one can conclude that no dust intrusion was occurring during the intensive field ~~campaign in Melpitz~~campaigns. Thus, the CCN concentration profiles were calculated following the continental aerosol branch in Mamouri and Ansmann (2016).

For this approach, the lidar-derived particle backscatter profiles are converted to extinction profiles by using a lidar ratio of 50 sr as a typical value for continental sites (Baars et al., 2017). The aerosol number concentration profiles for particles with a dry radius > 50 nm (n_{50}) are calculated using

$$n_{50,c,dry}(z) = c_{60,c} * \sigma_c^{X_c}(z)$$

with $c_{60,c}$=25.3 cm^{-3} and X_c=0.94 (see Mamouri and Ansmann (2016) for details). Finally, the CCN concentration at supersaturations $<$0.2 % is estimated by multiplying n_{50} with an enhancement factor of f = 1. The uncertainty of this estimation is at a factor of 2-3 according to Mamouri and Ansmann (2016).

3 Results

3.1 Composition of CCN

As described above, number concentrations of CCN over Germany for two time periods of the year 2013 have been calculated offline from aerosol particle number concentrations based on simulated mass concentrations of 7 different compounds: ammonium sulfate, ammonium nitrate, sulfate, organic and elemental carbon, sea salt and mineral dust. Similarly, representing a peak aerosol scenario over Europe, aerosol concentrations have been calculated for 1985 based on the simulations for the year 2013 (see section 2.2.2). Furthermore, the CCN parameterization has been applied to observed particle mass concentrations. The modeled CCN number concentrations were compared to ground-based in-situ measurements by a CCNC, and to vertical

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 two 2013 ~~campaign~~ campaigns and the ~~corrsponding period~~ corresponding periods 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~~In addition, the average ~~value~~ in-situ measured ~~by the CCNC was 1.1*10^9~~CCN number concentration is shown for comparison.

Data base / scenario	N_CCN$_{0.2\,\%}$, m^{-3}	AS	AN	SU	EC	OC	SS	DU
Modeled aerosol mass concentrations (1985)	~~5.2*10^9~~ 5.2×10^9	36	0	64	0	0.4	0.3	0.001
Modeled aerosol mass concentrations (2013)	~~9.4*10^8~~ 9.4×10^8	51	46	0.007	0	2.3	1.6	0.008
Measured aerosol mass concentrations (2013)	~~1.5*10^9~~ 1.5×10^9	35	53	0	0	7.4	0.3	4.0
Direct observation of CCN with CCNC (2013)	1.1×10^9							

profiles derived from lidar and helicopter-borne in-situ observations~~at Melpitz~~. Table 4 lists the total number concentration of CCN and the contribution of the individual compounds as average values for the simulated time period. Nowadays, the contribution of ammonium nitrate and ammonium sulfate are almost balanced~~, but~~. Due to the assumption that ammonium nitrate was not formed in the 1980s, ~~ammonium nitrate played almost no role~~there is no contribution from ammonium nitrate to CCN in the 1985 case. The concentration of ammonium sulfate in the atmosphere was far higher than today (see also section 2.2.2), resulting in almost no ammonia being available for the formation of ammonium nitrate. Instead, much more sulfuric acid could form during this time period ~~. Due to usage of these assumptions in the derivation of the 1985 aerosol mass concentrations, this effect can also be seen in the contributions to the CCN budget.~~

Comparing the two different methods of estimating todays CCN concentrations, differences can be seen especially for ammonium sulfate, organic carbon and mineral dust. The dust concentrations resulting from the gravimetrical methods are usually higher than simulated, because they result from the difference of the total gravimetric mass and the sum of the masses of the individual species and are not directly measured. This is why the error is quite large due to losses of the other species during the analytical processes. Furthermore, they may contain other undetected material than only mineral dust and also re-emitted soil dust, which is not included in the emission data used in the model simulations. The difference in ~~OC is probably~~ CCN from OC is partly due to the absence of secondary organic aerosol (SOA) in the model approach. SOA ~~contributes~~ generally can contribute a large fraction to the total concentration of organic aerosol mass (Jimenez et al., 2009) and also ~~Melpitz~~ at Melpitz SOA is known to ~~be a SOA dominated location~~ comprise a major fraction of the PM$_1$ aerosol (Poulain et al., 2011).

~~Figure~~ Fig. 2 shows the time series of ~~simulated CCN for~~ derived CCN from the model simulation (upper panel) and from gravimetrical aerosol measurements (lower panel) for both the spring and fall ~~periods in comparison with~~ period in comparison to the CCNC measurements at a supersaturation of 0.2 %. The same plot ~~is shown~~ for a supersaturation of 0.3~~% in the Supplemental Figure A1.The upper panel shows the results from the model simulations while the lower one shows the~~

~~results from parameterizing the gravimetrically determined particle mass concentrations. The good agreement between the CCN concentrations from~~ % is shown in Fig. A1 in the supplement. On average (see Tab. 4), CCN concentrations derived from modeled and observed aerosol mass deviate from the CCNC measurements by a factor of around 1.2 (16 % underestimation) and 1.4 (37 % overestimation), respectively. Taking into account the uncertainty due to assumptions in converting observed or modeled aerosol mass into number, the ~~direct measurements and the estimations from chemical analytics shows, that the~~ used CCN parametrization ~~works~~ is concluded to work reasonably well. Differences in the upper panels of Fig. 2 correspond to uncertainties in the actual aerosol simulation with the atmospheric transport model. Particularly in the first half of the spring episode, ~~nitrate~~ ammonium nitrate and ammonium sulfate concentrations and ~~also sulfate and~~ thus their contribution to the CCN number concentration were clearly underestimated. ~~In contrast, nitrate was overestimated by a factor of about 2~~ However, during the fall period the model often overestimates the concentration of ammonium sulfate and particularly ammonium nitrate (up to factor of 5) and hence the CCN concentrations. Deviations in ammonium nitrate might arise due to uncertainties of both modeling and observation. The emission of ammonia is depending on agricultural activity (e.g., manuring). Hence, the magnitude and timing of observed ammonium nitrate concentration peaks cannot be represented by the model, which uses monthly emission estimates. Since nitrate is volatile, high temperature within the sampling unit can lead to partial evaporation from the filters. An interesting episode occurred between day-of-year (doy) 255 and 257 (September 12-14, 2013) in the fall ~~episode~~period, resulting in clearly overestimated CCN ~~numbers~~number concentrations in the model. This was caused by a small surface low, which was centered above the measurement station on doy 255 and then moved eastward. The location of this surface low was not correctly simulated in the model and the corresponding precipitation and thus wet deposition of aerosol particles was missing, resulting in an overestimation of ~~particles. Furthermore, the photochemical reduction of nitrate was reduced due to cloudiness. A ridge of high pressure was following during the night of doy 257 to 258, which ended with a frontal passage and some precipitation, marking the end of this episode with clear overestimation of nitrate particles. Anyhow, since nitrate is problematic both to simulate (especially in spring) and to measure (especially in fall), the results compare satisfactorily well with the direct observation of the CCN number concentrations~~aerosol mass concentration and hence aerosol and CCN number concentration.

3.2 Comparison to in-situ CCN measurements

For a more evident comparison of the absolute number concentrations, Fig. 3 displays the ~~simulated~~ derived and measured CCN ~~numbers~~ number concentrations at a supersaturation of 0.2 % as a scatter plot for both episodes. As already seen in the time series plots in Fig. 2, the model ~~underestimates~~ tends to underestimate the CCN numbers ~~compared to both,~~ of the in-situ CCN measurements ~~and the CCN numbers derived from the gravimetric measurements~~ in the spring episode ~~.~~ (on average by 29 %). For the fall episode, an overestimation of 37 % was found (20 % without the outliers of the two days discussed above). In contrast, the CCN number concentration estimated from the ~~gravimetrical measured particle mass was higher than the direct CCN measurement. For the fall episode, a better agreement between model and observation was found, except for a few outliers during two days. As mentioned above, this is probably caused by the underestimation of precipitation associated with a small~~

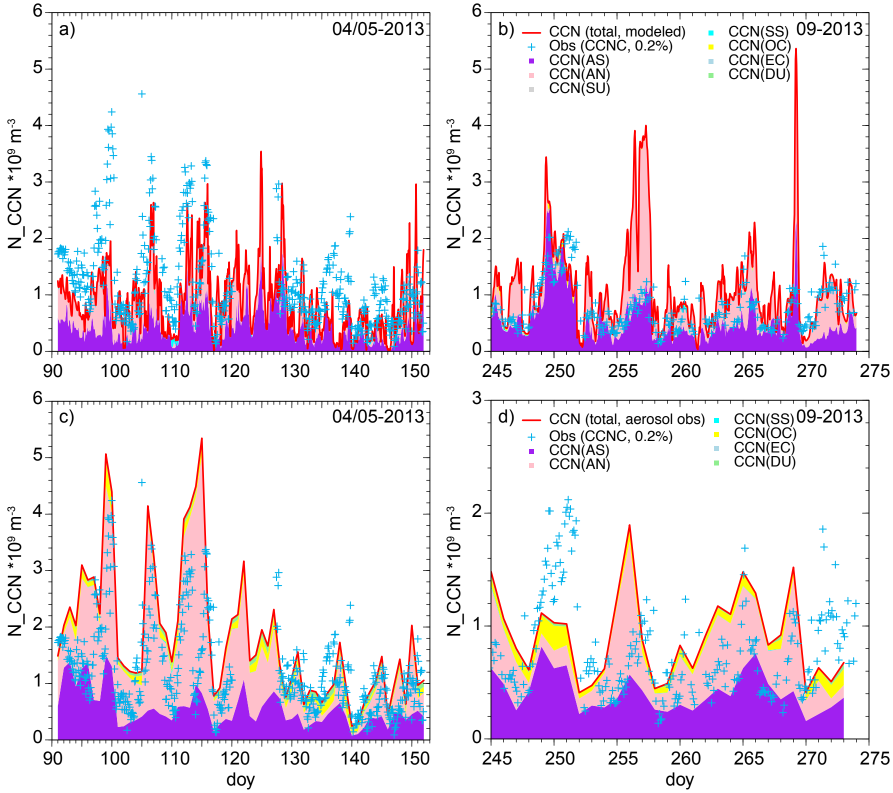

Figure 2. Simulated and measured CCN number concentrations in Melpitz at a supersaturation of 0.2 % during ~~HOPE-Jülich~~ the two HOPE campaigns (~~04/05 2013)~~ April to May and ~~HOPE-Melpitz (09/~~September 2013). The upper panel (a and b) shows the CCN number concentrations resulting from the simulated aerosol concentrations, the lower one (c and d) the CCN numbers resulting from measured aerosol concentrations using the same CCN parametrization. The colors represent the contributions to CCN of different species. The blue crosses indicate the CCN number concentrations using the CCNC. Please note the different time resolution for the observations, as well as the different scale for the CCN number concentration in plot d.

~~low pressure system in the simulation~~gravimetrically measured aerosol masses tends to overestimate the direct measurements in both periods (50 % in spring, 15 % in fall).

In Fig. 4 the ratio of the ~~potential CCN~~ number concentrations of CCN (N_CCN) and the total aerosol particles (N_CN) larger than a certain size is shown as comparison between simulation and observation. The upper panels display the fractions for a supersaturation of 0.2 % and particles larger than 110 nm for both episodes, the lower panels for a supersaturation of 0.3 % and particles larger than 80 nm, respectively. A ratio of exactly ~~one~~ 1.0 means, that as many particles would activate at the respective supersaturation as aerosol particles with a diameter larger than the threshold diameter of ~~110nm~~ 110 nm(N_CN$_{110\mathrm{nm}}$) and ~~80nm~~ 80 nm (N_CN$_{80\mathrm{nm}}$), respectively, are present in the atmosphere at this time. For the rural observation site Melpitz,

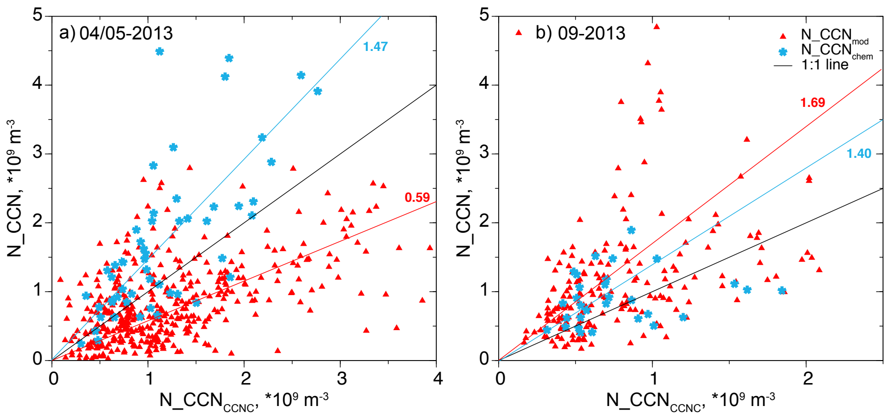

Figure 3. Comparison of ~~simulated~~ derived and measured CCN number concentrations in Melpitz at a supersaturation of 0.2%. Red triangles show results from the aerosol simulations, ~~green~~ blue stars ~~result~~ from applying the CCN parameterization to the ~~chemically~~ gravimetrically measured ~~particle~~ aerosol mass concentrations. The colored lines are the linear regressions. The slope of the fits are given at the regression lines.

this ratio is usually close to ~~one~~ 1.0 for 0.2 % and 110 nm~~and~~ , as well as 0.3 % and 80 nm (S. Henning, 2017, personal communication), ~~respectively,~~ which is why these two size threshold values were chosen~~for the displayed diagrams.~~

. The N_CCN~~$_{0.2\%}$~~$_{0.2\,\%}$ to N_CN$_{110\,\mathrm{nm}}$ ratios compare very well (on average 1.03 (observation) and 0.98 (model), respectively), but the model tends to overestimate the N_CCN~~$_{0.3\%}$~~$_{0.3\,\%}$ to N_CN$_{80\,\mathrm{nm}}$ ratios for both episodes (on average, 0.93 (observation) and 1.26 (model), respectively). This can be the result of the model either overestimating ~~potential CCN or underestimating~~ the CCN concentration or underestimating the aerosol particle number in the size range larger than 80 nm in diameter. ~~A too large number of CCN could result from too many large particles, which activate at lower supersaturations than in the real atmosphere, or overestimated particle hygroscopicity. Since κ is well documented in the literature, it is a less likely source of uncertainty. The detailed analysis in in~~ For both 0.2 % and 0.3 % supersaturation, the model underestimate the CCN concentration in total for both periods by 13 and 11 %, respectively (see also Figs. 2 and A1~~show that the CCN number concentration is in similar agreement with the observations for both supersaturations considered. However, as~~). The size distributions used to convert modeled aerosol mass to number were developed with data at Melpitz. Although they were shown able to represent the average total particle number concentration around 100 nm (Hande et al., 2016), uncertainty of the model estimate is due to the temporal variation of the aerosol size distribution and the composition of the aerosol particles. As can be seen ~~, uncertainties of the aerosol composition~~ in Figs. 2 and A1, these uncertainties can lead to up to a factor of 2 difference between CCN derived from modeled and observed aerosol masses. ~~On average, the model underestimates the CCN number by around 13% (cf. Fig. 3).~~

~~Due to the assumptions made to transfer modeled aerosol mass into number size distributions (see Tab. 1) the usually large number of particles in the Aitken or even nucleation range cannot be considered. As already shown by Hande et al. (2016), with this approach the number concentration of CN particles~~ Overall, the total particle number concentrations is in agreement to observations for particles larger than 110 nm ~~compares well with the measurements. On average, the model underestimates these numbers by less than~~ (10~~%~~ % underestimation). However, ~~this is different for smaller particle size ranges, e.g., the total number~~ the number concentration of particles larger than 80 nm is ~~underestimates~~ underestimated by 35~~% (Hande et al., 2016)~~ %. Hence, the underestimated modeled number concentration of aerosol particles in the size range between 80 and 110 nm in diameter is the main reason for the different behavior between the N_CCN~~$_{0.2\%}$~~$_{0.2\%}$ to N_CN$_{110\,\mathrm{nm}}$ ratio and the N_CCN~~$_{0.3\%}$~~$_{0.3\%}$ to N_CN$_{80\,\mathrm{nm}}$ ratio.

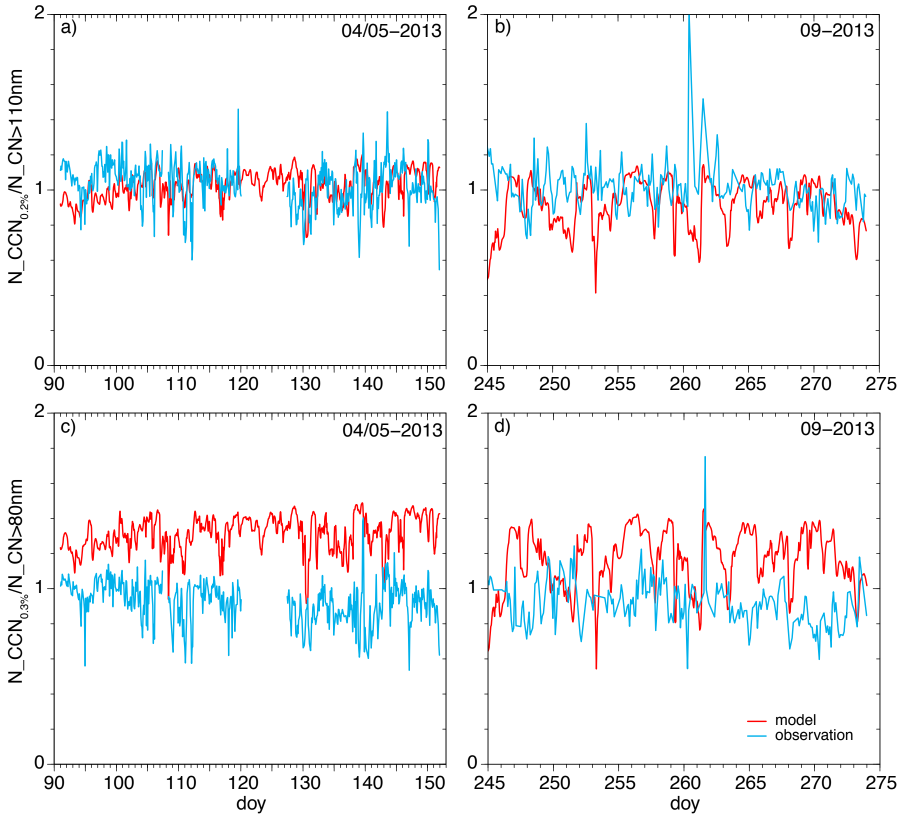

Figure 4. Comparison of the modeled and observed activated fraction (N_CCN / N_CN) at a supersaturation of 0.2 % (a and b) and 0.3 % (c and d), respectively. As number of total CN, the number concentration of CN > 110 nm (a and b) and > 80 nm (c and d), respectively, was used.

Fig. 5 shows the average N_CCN-to-N_CN ratio for five different supersaturations between 0.1 and 0.7 % for a cut-off diameter of 40 nm. It can be seen from this graph, that at a low supersaturation of 0.1 %, only very few particles activate, whereas almost all particles activate at a high supersaturation of 0.7 %. In the model, more of the available aerosol particles activate at ~~a~~ the respective supersaturation, which is most pronounced in the medium range of supersaturations. In this region the chemical composition and the assumptions for the size distributions of the particles are likely more important.

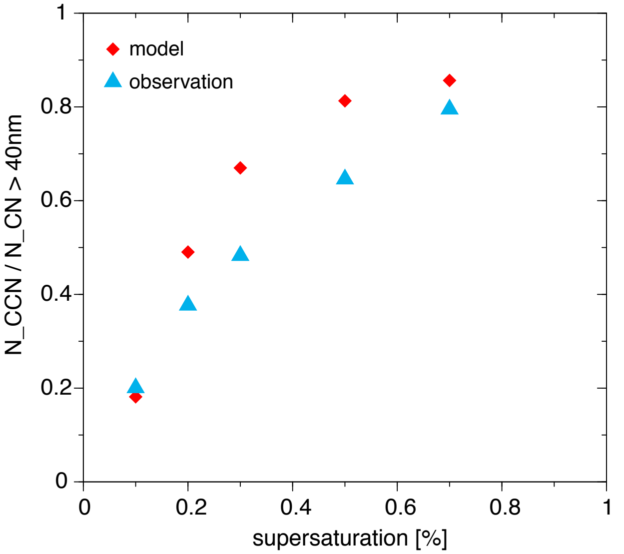

Figure 5. Simulated and observed fraction of ~~potential~~ CCN number concentration to the total ~~particle~~ number concentration of particles with a diameter larger than 40nm (N_CCN/N_CN) as a function of supersaturation.

3.3 Evaluation of the vertical structure of ~~CCNs~~CCN

In order to evaluate the vertical distribution of the CCN concentrations and investigate its change since the 1980s, the modeled vertical profiles are compared to measurements. ~~Figure 6 shows a comparison~~ Fig. 6 compares the simulated and observed vertical profiles of the CCN number concentration ~~vertical profiles over the two months spring episode in 2013 over Melpitz as simulated with COSMO-MSUCAT and~~ for the two periods in 2013. Fig. 6a shows the comparison to CCN derived from lidar ~~(6 a) and ACTOS in-situ (6b) observations~~ observation during the spring period at Jülich, and Fig. 6b the comparison to the in-situ observations by the helicopter-based platform ACTOS during the fall period at Melpitz. Displayed are the median values as well as the 0.25- and 0.75-quantiles. ~~Close~~ For the spring period and close to the ground, the ~~model compares quite well to the two observations. The~~ average CCN number concentration is overestimated by less than 50 %, which is in the range of the observation uncertainty of up to a factor of ~~2. At~~ 2-3. However, up to a height of ~~around~~ ~1.3 km~~the~~, marking the average height of the boundary layer, the overestimation increases up to a factor of ~2. Nevertheless, the displayed 0.25-0.75 quantile range still overlaps in the boundary layer. Above, the observed and measured CCN concentrations start to decrease considerably, but clearly ~~most~~ more strongly in the lidar observations. The model seems to transport too much aerosol mass into the free troposphere. In contrast to the model, the CCN number concentration derived from the lidar are on average negligible at heights above 4 km. Nevertheless, the variability of the observed CCN number concentrations is higher in the free troposphere. This is mainly an expression of ~~considerably~~ increased detection uncertainty. ~~Overall, the~~ The comparison to the in-situ observations by ACTOS during the fall period displayed in Fig. 6b reveals a stronger overestimation also close to the ground by a factor of ~2. Also for this comparison, the modeled CCN number concentration does not as strongly decrease with height above the boundary layer (~1.5 km), hence increasing the overestimation. Note, that the larger variability of the median with height and the smaller 0.25-0.75 quantile range is caused by the smaller sample size of only 8 distinct cases compared to the 48 days with several hours of lidar observations during the spring period. Furthermore, the ACTOS observation have a general uncertainty of only ~10 %. This, therefore, manifests the tendency of the model to overestimate the average CCN concentrations in the boundary layer by up to a factor of 2 and higher above the boundary layer. The general overestimation could be reduced by assuming different aerosol size distributions, which are used to convert modeled aerosol mass into number. However, the utilized size distributions were derived from data at Melpitz and any other size distribution would therefore be less justified. It can be expected that the size distribution is not constant in time and in space as currently applied. Simulations that treat the aerosol in a size-resolved manner including aerosol microphysics are a useful tool to provide more insight into the temporal and spatial variability of the aerosol size distribution and hence the CCN number concentration. However, due to the increased degrees of freedom and similar assumptions, such as the size distribution during the emission, the results are not necessarily more accurate. Overall, although the model tends to overestimate the average CCN concentrations, the modeled present day-CCN number concentration is in-line with the observations, whereas the estimated profile for the 1980s is far outside today's observational range (cf. Figs. 6 and ~~??~~7). This ~~clearly shows~~ indicates the influence of anthropogenic air pollution on the CCN number.

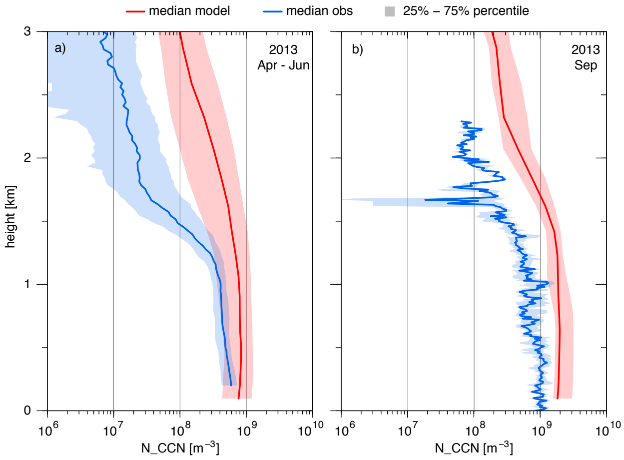

Figure 6. Comparison of the simulated vertical profiles of CCN number concentration ~~vertical profiles~~ (red) to profiles derived from observations (blue) of (a) lidar (04/05 2013) at Jülich, Germany, and (b) ACTOS (09/2013) ~~over~~ at Melpitz, Germany. The CCN number concentrations were calculated or measured for a supersaturation of 0.2 %. The shading depicts the range between the 0.25- and the 0.75-quantile. On 48 and 8 different days, 335 and 27 model profiles (instantaneous hourly output), which matched the time of observations, could be taken into account for the spring and fall period.

3.4 Present day and historic vertical CCN profiles

For each of the two ~~evaluation periods~~ periods and the full domain, a temporally and spatially averaged vertical profile of the CCN concentration was calculated for the year 2013 and the year 1985 emission scenario, which is displayed together with the 0.05, 0.25, 0.75 and 0.95 quantiles in Fig. 7a - d. For the calculation, a vertical velocity of $1\,\mathrm{ms^{-1}}$ was assumed. Hence, in contrast to the previous analysis the supersaturation depends on the aerosol composition and varies spatially and temporally. The shape and values of the profiles show no major differences for the spring and fall episode. Close to the ground, where aerosol particles are emitted, the number concentrations of ~~potential~~ CCN are higher than in the free troposphere. With increasing height, the number of aerosol particles and thus also that of ~~potential~~ CCN is decreasing. This is the case for both the 2013 and 1985 scenario. In 2013, the concentrations are almost constant up to a height of $1\,\mathrm{km}$ (around ~~$1.0*10^9$~~ $1.0\times10^9\,\mathrm{m^{-3}}$) due to the well mixed boundary layer and decrease above (Fig. 7a, d). This is less pronounced in the year 1985 simulations (Fig. 7c, d), in which the concentrations close to the ground are much higher (around ~~$3*10^9$~~ $3\times10^9\,\mathrm{m^{-3}}$) and decrease almost immediately with height. At the top of the uppermost simulated layer ($8\,\mathrm{km}$), similar concentrations of ~~$5*10^7$ to $1*10^8$~~ 5×10^7 to $1\times10^8\,\mathrm{m^{-3}}$ were found for both, the present day and peak aerosol scenario. Due to different aerosol composition and, hence, aerosol hygroscopicity between 1985 and 2013, the shape of the CCN profiles in the two scenarios differs. Based on the CCN profiles, a scaling factor for the CCN concentration was calculated, which varies with height (Fig.~~7 e~~ 7e, f). This scaling factor describes the mean temporal trend of the CCN number concentration between past peak aerosol in the 1980s and present day conditions in Europe and ~~can easily be used~~ are useful for sensitivity studies. The difference in the height dependency of the number concentrations between the 2013 and 1985 simulations is the reason for the ~~bend in the~~ curvature in the plot of the scaling factor at around $1\,\mathrm{km}$ height (Fig. 7e, f), because at this height, also the concentrations in the 2013 simulations start to decrease. Close to the ground, a factor of around two was found. The efficacy of pollution reduction policies and the breakdown of industrial production in ~~Eastern Europe since the~~ the former East-bloc countries at the end of the 1980s becomes evident, in relative terms, most pronounced in the height between 2 and $5\,\mathrm{km}$, where a scaling factor of up to a factor of 3.5 was found. In the upper troposphere, the scaling factor decreases to around one, which means there is no difference between the 1980s and present day concentrations.

4 Summary and conclusions

The CCN number concentrations from different simulation estimates and observation techniques were compared ~~in this study for the period~~ for two periods of the HOPE field ~~experiment~~ experiments in Germany in spring and fall 2013. Based on simulations of the mass concentrations of different aerosol species (ammonium sulfate, ammonium nitrate, sulfate, organic carbon, elemental carbon, sea salt, and mineral dust) using the regional ~~CTM~~ chemistry-transport model COSMO-MUSCAT, the CCN number was ~~parametrized~~ computed offline using a state-of-the-art parameterization for cloud droplet activation. The resulting CCN number concentrations were compared to ~~(i)~~ direct CCN measurements with a CCN counter, ~~(ii)~~ CCN number concentrations derived from applying the activation parameterization to gravimetrically measured aerosol mass concentrations, and ~~(iii) to~~ vertical profiles derived from lidar observations and helicopter-borne in-situ measurements. In addition, CCN number con-

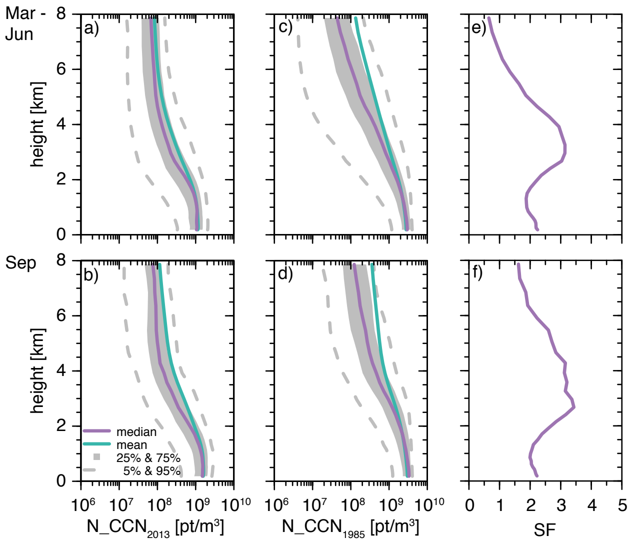

Figure 7. Spatial and temporal averaged vertical profile of the CCN number concentration as computed by COSMO-MUSCAT for the spring and fall period in 2013 (a and b), the estimation for the respective 1985 peak aerosol scenario (c and d) and scaling factor (SF) for the two scenarios (SF=N_CCN$_{1985}$ / N_CCN$_{2013}$; e and f). For the calculation of the CCN number concentration, a vertical velocity of 1 ms^{-1} was assumed.

centrations for the corresponding periods in the year 1985 were computed based on the COSMO-MUSCAT simulations ~~for the corresponding period in the year 1985, when industrial~~ of the year 2013, as exemplary year for the 1980s when anthropogenic air pollution in Central Europe had peaked. Comparing the ~~year~~ results for the years 2013 and 1985 ~~results~~ allows to investigate the impact of anthropogenic air pollution and the potential of reduction measures on the atmospheric CCN budget.

~~The quality of the modeled CCN number concentrations is defined by both, the quality of the aerosol particle simulation and the CCN parametrization. From the good agreement between CCN derived from gravimetrical measurementsand CCNC measurements, it can be concluded that the cloud droplet activation and growth parameterization gives reasonable results. Discrepancies of the offline CCN calculation from the model simulations can then be concluded resulting~~ At the ground and averaged over the full investigation period, the model-derived CCN concentration (for a supersaturation of 0.2 %) were about 16 % lower than the directly measured CCN concentrations and 37 % lower than the CCN concentrations derived from aerosol mass measurements. Hence, model and observation agree well for the longterm average. However, the deviations were different for the individual periods with 29 % underestimation of the measured CCN concentrations by the model in the spring period and 37 % overestimation for the fall period. Discrepancies between observed and modeled CCN concentrations likely resulted mostly from uncertainties in the modeled aerosol mass and composition as well as the assumptions for the ~~transfer~~ conversion from particle mass into number size ~~distribution.~~distributions, which do not allow for the necessary flexibility to consider weather and tranport-related heterogeity.. The comparison of the ratio of the CCN number concentration and the total particle number of particles larger than 110~~-~~ nm in diameter shows a good agreement between model and observation for 0.2~~-~~ % supersaturation. However, for supersaturations between 0.2 % and 0.7 % and smaller threshold sizes to define CN (e.g., particles larger than 40 nm), the model overestimates the activated particle fraction. Since the assumed ~~size distributions focus on the correct prediction~~ prescribed size distributions were developed to correctly predict the average number of accumulation mode particles, which are the most relevant for deriving CCN number concentrations, the number of particles smaller than ∼100 nm is very likely underrepresented. As a non-linear process, aerosol activation depends strongly on the current ambient aerosol size distribution, which can vary considerably both temporally and spatially. Hence, the application of fixed size distributions in order to convert modeled aerosol mass to number concentrations is a source of uncertainty, which only for longterm averages might cancel out.

~~The vertical structure of the simulated CCN number concentration was also shown to agree reasonably well with ground-based remote sensing and airborne in-situ measurements, even though the variability could not be reproduced by the model. Close to the ground, model and observation agree well, but the measurement based profiles show a much larger range, which is probably due to both, a high variability in the real atmosphere during the two months of~~ At the measurement station, the ~~experiment and measurement uncertainties (factor 2-3). In conclusion~~model-derived average CCN concentration for the year 1985 was more than 5 times higher than for the year 2013. Again, the application of fixed prescribed parameters for the number size distributions likely is a source of uncertainty since the aerosol size distribution in 2013 and 1985 were not necessarily similar.

Within the boundary layer, the simulated vertical profiles of the present-day ~~simulation~~ CCN concentration are within the variability range of the ~~measurement-based profiles and thus represent realistic conditions. The~~ CCN derived from lidar measurements but do deviate from the in-situ helicopter-borne CCN measurements outside their 0.25-0.75 quantile range (and up to a factor of 2 for the median). The strong decrease of the observed CCN concentrations above the boundary layer could not be met by the model, hence strongly overestimating the CCN concentration in the free troposphere. The 1985 simulation, however, has much larger CCN number concentration far outside the variability range of the present-day observations~~, as expected. A vertical-resolved scaling factor between the~~.

By comparing the CCN concentrations modeled for the year 2013 and 1985, the effect of strict emission reduction policies and reorganization of industrial production in Eastern Europe after 1990 becomes apparent. A domain and time averaged vertically resolved scaling factor for the CCN concentration between the year 2013 and year 1985 ~~results~~ was computed, which is well suited for application in model sensitivity studies.

The scaling factor for estimating ~~peak aerosol~~ the CCN concentrations during the 1980s from current simulations is not vertically homogeneous. Close to the ground, a scaling factor of 2 was determined, increasing to 3.5 between 2 and 5 km height. ~~Here the effect of strict emission reduction policies and reduction in industrial production in Eastern Europe especially in the 1990s becomes apparent.~~ Towards the upper troposphere at around 8 km height, the scaling factor decreases again to 1. This means, the dynamics and thermodynamics of the troposphere have a large influence on the distribution of the aerosol particles and thus the CCN distribution. Especially the height range of ~~2~~ up to 5 km, where a very high CCN number concentration during the 1980s was found, is important for cloud and precipitation formation in the mid-latitudes. A significantly higher number of CCN points to large differences in the cloud droplet number concentration and thus the radiative properties of the clouds as well as ~~in~~ the precipitation probability during that time. The analysis of the radiative impacts including effects on cloud cover and albedo effects ~~are~~ should be subject of future studies.

Data availability. Data used in this manuscript can be provided upon request by email to the corresponding author, Christa Genz (christa.genz@idiv.de).

Appendix A

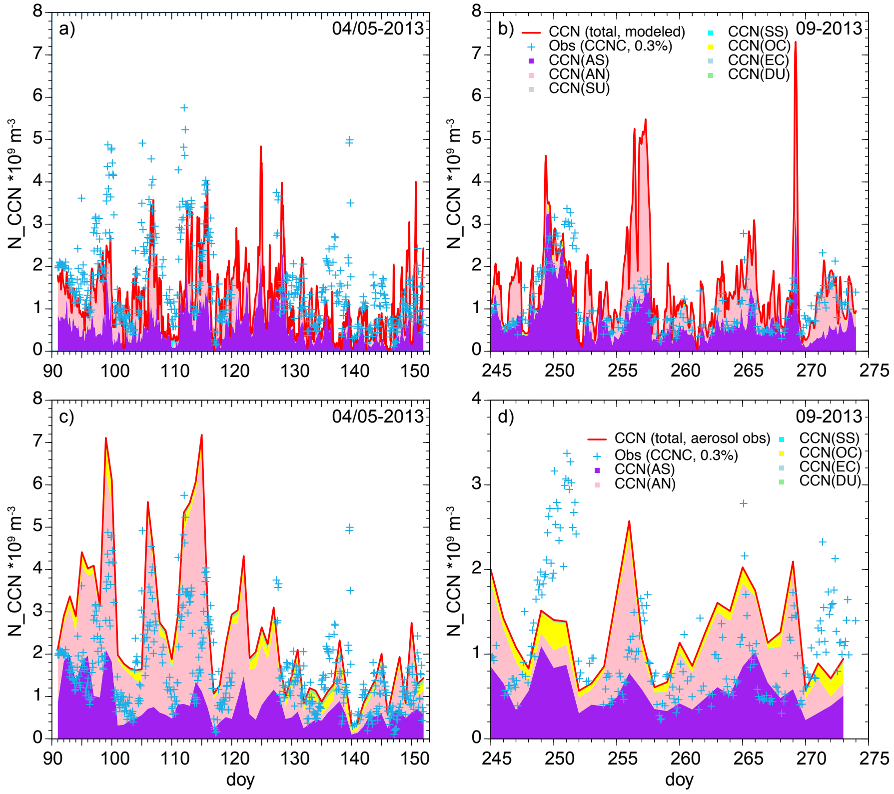

Figure A1. Simulated and measured CCN number concentrations in Melpitz at a supersaturation of 0.3 % during ~~HOPE-Jülich~~ the two HOPE campaigns (~~04/05 2013)~~ April to May and ~~HOPE-Melpitz (09/~~September 2013). The upper panel (a and b) shows the CCN number concentrations resulting from the simulated aerosol concentrations, the lower one (c and d) the CCN numbers resulting from measured aerosol concentrations using the same CCN parametrization. The colors represent the contributions to CCN of different species. The blue crosses indicate the CCN number concentrations using the CCNC. Please note the different time resolution for the observations, as well as the different scale for the CCN number concentration in plot d.

Author contributions. Christa Genz working with Ina Tegen and Bernd Heinold ran the COSMO-MUSCAT model, and performed the aerosol evaluation and CCN concentration calculation. Roland Schrödner joined during the analysis and coordinated the revision. Silvia Henning provided the CCNC measurements, Holger Baars the lidar derived CCN profiles and Gerald Spindler obtained the chemical measurements and analysis. Christa Engler and Roland Schrödner prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This work was funded by the Federal Ministry of Education and Research in Germany (BMBF) through the research programme "High Definition Clouds and Precipitation for Climate Prediction - HD(CP)^2" (FKZ: 01LK1503F, 01LK1502I, 01LK1209C and 01LK1212C). The authors wish to thank Luke Hande for providing a version of CCN-calculation code, which served as basis for the estimations shown here. We also acknowledge good cooperation and support from the German Weather Service (Deutscher Wetterdienst, DWD) and the high performance computing center Jülich. Furthermore the authors acknowledge support from ACTRIS under grant agreement no. 262 254 of the European Union Seventh Framework Programme (FP7/2007-2013).

References

Abdul-Razzak, H. and Ghan, S.: A parametreization of aerosol activation: 2. Multiple aerosol types, Journal of Geophysical Research, 105, 2000.

Abdul-Razzak, H., Ghan, S., and Rivera-Carpio, C.: A parametreization of aerosol activation: 1. Single aerosol types, Journal of Geophysical Research, 103, 6123–6131, 1998.

Baars, H., Kanitz, T., Engelmann, R., Althausen, D., Heese, B., Komppula, M., Preißler, J., Tesche, M., Ansmann, A., Wandinger, U., Lim, J.-H., Ahn, J., Stachlewska, I., V, A., Marinou, E., Seifert, P., Hofer, J., Skupin, A., Schneider, F., Bohlmann, S., Foth, A., Bley, S., Pfüller, A., Giannakaki, E., Lihavainen, H., Viisanen, Y., Hooda, R., Pereira, S., Bortoli, D., Wagner, F., Mattis, I., Janicka, L., Markowicz, K., Achtert, P., Artaxo, P., Pauliquevis, T., Souza, R., van Zyl, V. S. P., Beukes, J., Sun, J., Rohwer, E., Deng, R., Mamouri, R.-E., and Zamorano, F.: An overview of the first decade of Polly^{NET}: an emerging network of automated Raman-polarization lidars for continuous aerosol profiling, Atmospheric Chemistry and Physics, 16, 5111–5137, 2016.

Baars, H., Seifert, P., Engelmann, R., and Wandinger, U.: Target categorization of aerosol and clouds by continuous multiwavelength-polarization lidar measurements, Atmospheric Measurement Techniques, 10, 3175–3201, https://doi.org/10.5194/amt-10-3175-2017, 2017.

Bangert, M., Kottmeier, C., Vogel, B., and Vogel, H.: Regional scale effects of the aerosol cloud interaction simulated with an online coupled comprehensive chemistry model, Atmospheric Chemistry and Physics, 11, 4411–4423, https://doi.org/10.5194/acp-11-4411-2011, 2011.

Bègue, N., Tulet, P., Pelon, J., Aouizerats, B., Berger, A., and Schwarzenboeck, A.: Aerosol processing and CCN formation of an intense Saharan dust plume during the EUCAARI 2008 campaign, Atmospheric Chemistry and Physics, 15, 3497–3516, https://doi.org/10.5194/acp-15-3497-2015, https://www.atmos-chem-phys.net/15/3497/2015/, 2015.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S., Sherwood, S., X., B. S., and Zhang: Clouds and Aerosols, in: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge Univ Press, New York, 2013.

Bühl, J., Seifert, P., Wandinger, U., Baars, H., Kanitz, T., Schmidt, J., Myagkov, A., Engelmann, R., Skupin, A., Heese, B., Klepel, A., Althausen, D., and Ansmann, A.: LACROS: the Leipzig Aerosol and Cloud Remote Observations System, in: Remote Sensing of Clouds and the Atmosphere XVIII; and Optics in Atmospheric Propagation and Adaptive Systems XVI, edited by Comeron, A., Kassianov, E. I., Schäfer, K., Stein, K., and Gonglewski, J. D., vol. 8890, pp. 1 – 6, International Society for Optics and Photonics, SPIE, https://doi.org/10.1117/12.2030911, https://doi.org/10.1117/12.2030911, 2013.

Cherian, R., Quaas, J., Salzmann, M., and Wild, M.: Pollution trends over Europe constrain global aerosol forcing as simulated by climate models, Geophysical Research Letters, 41, 2176–2181, https://doi.org/10.1002/2013GL058715, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1002/2013GL058715, 2014.

Costa-Surós, M., Sourdeval, O., Acquistapace, C., Baars, H., Carbajal Henken, C., Genz, C., Hesemann, J., Jimenez, C., König, M., Kretzschmar, J., Madenach, N., Meyer, C. I., Schrödner, R., Seifert, P., Senf, F., Brueck, M., Cioni, G., Engels, J. F., Fieg, K., Gorges, K., Heinze, R., Siligam, P. K., Burkhardt, U., Crewell, S., Hoose, C., Seifert, A., Tegen, I., and Quaas, J.: Detection and attribution of aerosol-cloud interactions in large-domain large-eddy simulations with ICON, Atmospheric Chemistry and Physics Discussions, 2019, 1–29, https://doi.org/10.5194/acp-2019-850, https://www.atmos-chem-phys-discuss.net/acp-2019-850/, 2019.

Dipankar, A., Stevens, B., Heinze, R., Moseley, C., Zängl, G., Giorgetta, M., and Brdar, S.: Large eddy simulation using the general circulation model ICON, Journal of Advances in Modeling Earth Systems, 7, 963–986, 2015.

Duplissy, J., DeCarlo, P., Dommen, J., Alfarra, M., Metzger, A., Barmpadimos, I., Prevot, A., Weingartner, E., Tritscher, T., Gysel, M., Aiken, A., Jimenez, J., Canagaratna, M., Worsnop, D., Collins, D., Tomlinson, J., and Baltensperger, U.: Relating hygroscopicity and composition of organic aerosol particulate matter, Atmospheric Chemistry and Physics, 11, 1155–1165, 2011.

Düsing, S., Wehner, B., Seifert, P., Ansmann, A., Baars, H., Ditas, F., Henning, S., Ma, N., Poulain, L., Siebert, H., Wiedensohler, A., and Macke, A.: Helicopter-borne observations of the continental background aerosol in combination with remote sensing and ground-based measurements, Atmospheric Chemistry and Physics, 18, 1263–1290, https://doi.org/10.5194/acp-18-1263-2018, https://www.atmos-chem-phys.net/18/1263/2018/, 2018.

EMEP: European Monitoring and Evaluation Programme, http://www.emep.int/, 2009.

Engelmann, R., Kanitz, T., Baars, H., Heese, B., Althausen, D., Skupin, A., Wandinger, U., Komppula, M., Stachlewska, I., Amiridis, V., Marinou, E., Mattis, I., Linné, H., and Ansmann, A.: The automated multiwavelength Raman polarization and water-vapor lidar Polly^{XT}: the neXT generation, Atmospheric Measurement Techniques, 9, 1767–1784, 2016.

Engler, C., Rose, D., Wehner, B., Wiedensohler, A., Brüggemann, E., Gnauk, T., Spindler, G., Tuch, T., and Birmili, W.: Size distributions of non-volatile particle residuals (Dp < 800nm) at a rural site in Germany and relation to air mass origin, Atmospheric Chemistry and Physics, 7, 5785–5802, 2007.

Fanourgakis, G. S., Kanakidou, M., Nenes, A., Bauer, S. E., Bergman, T., Carslaw, K. S., Grini, A., Hamilton, D. S., Johnson, J. S., Karydis, V. A., Kirkevåg, A., Kodros, J. K., Lohmann, U., Luo, G., Makkonen, R., Matsui, H., Neubauer, D., Pierce, J. R., Schmale, J., Stier, P., Tsigaridis, K., van Noije, T., Wang, H., Watson-Parris, D., Westervelt, D. M., Yang, Y., Yoshioka, M., Daskalakis, N., Decesari, S., Gysel-Beer, M., Kalivitis, N., Liu, X., Mahowald, N. M., Myriokefalitakis, S., Schrödner, R., Sfakianaki, M., Tsimpidi, A. P., Wu, M., and Yu, F.: Evaluation of global simulations of aerosol particle and cloud condensation nuclei number, with implications for cloud droplet formation, Atmospheric Chemistry and Physics, 19, 8591–8617, https://doi.org/10.5194/acp-19-8591-2019, https://www.atmos-chem-phys.net/19/8591/2019/, 2019.

Friedman, B., Zelenyuk, A., Beranek, J., Kulkarni, G., Pekour, M., Hallar, A. G., McCubbin, I., Thornton, J., and Cziczo, D.: Aerosol measurements at a high-elevation site: composition, size, and cloud condensation nuclei activity, Atmospheric Chemistry and Physics, 13, 11 839–11 851, https://doi.org/10.5194/acp-13-11839-2013, 2013.

Ghan, S., Laulainen, N., Easter, R., Wagener, R., Nemesure, S., Chapman, E., Zhang, Y., and Leung, R.: Evaluation of aerosol direct radiative forcing in MIRAGE, Journal of Geophysical Research, 106, 5295–5316, 2001.

Ghan, S., Rissman, T., Elleman, R., Ferrare, R., Turner, D., Flynn, C., Wang, J., Ogren, J., Hudson, J., Jonsson, H., VanReken, T., Flagan, R., and Seinfeld, J.: Use of in situ cloud condensation nuclei, extinction, and aerosol size distribution measurements to test a method for retrieving cloud condensation nuclei profiles from surface measurements, Journal of Geophysical Research, 111, D05S10, https://doi.org/10.1029/2004JD005752, 2006.

Gysel, M. and Stratmann, F.: WP3 - NA3: In-situ chemical, physical and optical properties of aerosols, http://www.actris.net/Publications/ACTRISQualityStandards/tabid/11271/language/en-GB/Default.aspx, 2013.

Hammer, E., Bukowiecki, N., Gysel, M., Jurányi, Z., Hoyle, C., Vogt, R., Baltensperger, U., and Weingartner, E.: Investigation of the effective peak supersaturation for liquid-phase clouds at the high-alpine site Jungfraujoch, Switzerland (3580ma.s.l.), Atmospheric Chemistry and Physics, 14, 1123–1139, https://doi.org/10.5194/acp-14-1123-2014, 2014.

Hande, L., Engler, C., Hoose, C., and Tegen, I.: Parameterizing cloud condensation nuclei concentrations during HOPE, Atmospheric Chemistry and Physics, 16, 12 059–12 079, 2016.

Hausmann, K.: personal communication with Kevin Hausmann from Umweltbundesamt (German Federal Environmental Agency, UBA), 2017.

Heinold, B., Tegen, I., Schepanski, K., Tesche, M., Esselborn, M., Freudenthaler, V., Gross, S., Kandler, K., Knippertz, P., and Müller, D.: Regional modeling of Saharan dust and biomass-burning smoke, Tellus B, 63, 781–799, 2011.

Heinze, R., Moseley, C., Böske, L., Muppa, S., Maurer, V., Raasch, S., and Stevens, B.: Evaluation of large-eddy simulations forced with mesoscale model output for a multi-week period during a measurement campaign, Atmospheric Chemistry and Physics, 17, 7083–7109, 2017.

Henning, S., Wex, H., Hennig, T., Kiselev, A., Snider, J., Rose, D., Dusek, U., Frank, G., Pöschl, U., Kristensson, A., Bilde, M., Tillmann, R., Kiendler-Scharr, A., Mentel, T., Walter, S., Schneider, J., Wennrich, C., and Stratmann, F.: Soluble mass, hygroscopic growth, and droplet activation of coated soot particles during LACIS Experiment in November (LExNo), Journal of Geophysical Research, 115, doi: 10.1029/2009JD012 626, 2010.

Henning, S., Dieckmann, K., Ignatius, K., Schäfer, M., Zedler, P., Harris, E., Sinha, B., van Pinxteren, D., Mertes, S., Birmili, W., Merkel, M., Wu, Z., Wiedensohler, A., Wex, H., Herrmann, H., and Stratmann, F.: Influence of cloud processing on CCN activation behaviour in the Thuringian Forest, Germany during HCCT-2010, Atmospheric Chemistry and Physics, 14, 7859–7868, https://doi.org/doi:10.5194/acp-14-7859-2014, 2014.

Hill, P., Morcrette, C., and Boutle, I.: A regime-dependent parameterization of subgrid-scale cloud water content variability, Quarterly Journal of the Royal Meteorological Society, 141, 1975–1986, 2015.

Hinneburg, D., Renner, E., and Wolke, R.: Formation of secondary inorganic aerosols by power plant emissions exhausted through cooling towers in Saxony, Environmental Science and Pollution Research, 16, 25–35, https://doi.org/10.1007/s11356-008-0081-5, https://doi.org/10.1007/s11356-008-0081-5, 2009.

Jimenez, J., Canagaratna, M., Donahue, N., Prevot, A., Zhang, Q., Kroll, J., DeCarlo, P., Allan, J., Coe, H., Ng, N., Aiken, A., Docherty, K., Ulbrich, I., Grieshop, A., Robinson, A., Duplissy, J., Smith, J., Wilson, K., Lanz, V., Hueglin, C., Sun, Y., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J., Collins, D., Cubison, M., Dunlea, E., Huffman, J., Onasch, T., Alfarra, M., Williams, P., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J., Zhang, Y., Dzepina, K., Kimmel, J., Sueper, D., Jayne, J., Herndon, S., Trimborn, A., Williams, L., Wood, E., Middlebrook, A., Kolb, C., Baltensperger, U., and Worsnop, D.: Evolution of Organic Aerosols in the Atmosphere, Science, 326, 1525–1529, 2009.

Köhler, H.: The nucleus in and the growth of hygroscopic droplets, Transactions of the Faraday Society, 32, 1152–1161, 1936.

Macke, A., Seifert, P., Baars, H., Barthlott, C., Beekmans, C., Behrendt, A., Bohn, B., Brueck, M., Bühl, J., Crewell, S., Damian, T., Deneke, H., Düsing, S., Foth, A., Girolamo, P. D., Hammann, E., Heinze, R., Hirsikko, A., Kalisch, J., Kalthoff, N., Kinne, S., Kohler, M., Löhnert, U., Madhavan, B. L., Maurer, V., Muppa, S. K., Schween, J., Serikov, I., Siebert, H., Simmer, C., Späth, F., Steinke, S., Träumner, K., Trömel, S., Wehner, B., Wiesner, A., Wulfmeyer, V., and Xie, X.: The HD(CP)^2 Observational Prototype Experiment (HOPE) - an overview, Atmospheric Chemistry and Physics, 17, 4887–4914, 2017.

Majewski, D., Liermann, D., Prohl, P., Ritter, B., Buchhold, M., Hanisch, T., Paul, G., Wergen, W., and Baumgardner, J.: The Operational Global Icosahedral–Hexagonal Gridpoint Model GME: Description and High-Resolution Tests, Monthly Weather Review, 130, 319–338, https://doi.org/10.1175/1520-0493(2002)130<0319:TOGIHG>2.0.CO;2, https://doi.org/10.1175/1520-0493(2002)130<0319:TOGIHG>2.0.CO;2, 2002.

Mamouri, R. E. and Ansmann, A.: Potential of polarization lidar to provide profiles of CCN- and INP-relevant aerosol parameters, Atmospheric Chemistry and Physics, 16, 5905–5931, 2016.

Niedermeier, D., Wex, H., Voigtländer, J., Stratmann, F., Brüggemann, E., Kiselev, A., Henk, H., and Heintzenberg, J.: LACIS-measurements and parameterization of sea-salt particle hygroscopic growth and activation, Atmospheric Chemistry and Physics, 8, 579–590, 2008.

Petters, M. and Kreidenweis, S.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmospheric Chemistry and Physics, 7, 1961–1971, 2007.

Poulain, L., Spindler, G., Birmili, W., Plass-Dülmer, C., Wiedensohler, A., and Herrmann, H.: Seasonal and diurnal variations of particulate nitrate and organic matter at the IfT research station Melpitz, Atmospheric Chemistry and Physics, 11, 12 579–12 599, https://doi.org/10.5194/acp-11-12579-2011, 2011.

Roberts, G. and Nenes, A.: A continuous-flow streamwise thermal-gradient CCN chamber for atmospheric measurements, Aerosol Science and Technology, 39, 206–221, 2005.

Schmale, J., Henning, S., Henzing, B., Keskinen, H., Sellegri, K., Ovadnevaite, J., Bougiatioti, A., Kalivitis, N., Stavroulas, L., Jefferson, A., Park, M., Schlag, P., Kristensson, A., Iwamotol, Y., Pringle, K., Reddington, C., Aalto, P., Aijala, M., Baltensperger, U., Bialek, J., Birmili, W., Bukowiecki, N., Ehn, M., Fjaeraa, A., Fiebig, M., Frank, G., Frohlich, R., Frumau, A., Furuyals, M., E. Hammerl', E. H., Heikkinen, L., Herrmann, E., Holzinger, R., Hyonols, H., Kanakidoug, M., Kiendler-Scharr, A., Kinouchi, K., Kos, G., Kulmala, M., Mihalopoulos, N., Motos, G., Nenes, A., O'Dowd, C., Paramonov, M., Petaja, T., Picard, D., Poulain, L., Prevot, A., Slowik, J., Sonntag, A., Swietlicki, E., Svenningsson, B., Tsurumaru, H., Wiedensohler, A., Wittbom, C., Ogren, J., Matsuki, A., Yum, S., Myhre, C., Carslaw, K., Stratmann, F., and Gysel, M.: Data Descriptor: Collocated observations of cloud condensation nuclei, particle size distributions, and chemical composition, Scientific Data, 4, https://doi.org/10.1038/sdata.2017.3, 2017.

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

Seifert, A. and Beheng, K.: A two-moment cloud microphysics parameterization for mixed-phase clouds. Part 1: Model description, Meteorology and Atmospheric Physics, 92, 45–66, https://doi.org/10.1007/s00703-005-0112-4, 2006.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics, Johl Wiley & Sons, Inc., New York, Chichester, Weinheim, Brisbane, Singapore, Toronto, 1326pp, 1998.

Siebert, H., Franke, H., Lehmann, K., Maser, R., Saw, E. W., Schell, D., Shaw, R. A., and Wendisch, M.: Probing Finescale Dynamics and Microphysics of Clouds with Helicopter-Borne Measurements, Bulletin of the American Meteorological Society, 87, 1727–1738, https://doi.org/10.1175/BAMS-87-12-1727, 2006.

Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J.-K.: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe - PART I: Unified EMEP Model Description, Norwegian Meteorological Institute, emep status report 2003 edn., 2003.

Smith, S. J., van Aardenne, J., Klimont, Z., Andres, R. J., Volke, A., and Delgado Arias, S.: Anthropogenic sulfur dioxide emissions: 1850 - 2005, Atmospheric Chemistry and Physics, 11, 1101–1116, https://doi.org/10.5194/acp-11-1101-2011, https://www.atmos-chem-phys.net/11/1101/2011/, 2011.

Spindler, G., Gnauk, T., Grüner, A., Iinuma, Y., Müller, K., Scheinhardt, S., and Herrmann, H.: Size-segregated characterization of PM_{10} at the EMEP site Melpitz (Germany) using a five-stage impactor: a six year study, Journal of Atmospheric Chemistry, 69, 127–157, 2012.

Spindler, G., Grüner, A., Müller, K., Schlimper, S., and Herrmann, H.: Long-term size-segregated particle (PM_{10}, $PM_{2.5}$, PM_1) characterization study at Melpitz - influence of air mass inflow, weather condition and season, Journal of Atmospheric Chemistry, 70, 165–195, 2013.

Spracklen, D. V., Carslaw, K. S., Pöschl, U., Rap, A., and Forster, P. M.: Global cloud condensation nuclei influenced by carbonaceous combustion aerosol, Atmospheric Chemistry and Physics, 11, 9067–9087, https://doi.org/10.5194/acp-11-9067-2011, https://www.atmos-chem-phys.net/11/9067/2011/, 2011.

Sudhakar, D., Quaas, J., Wolke, R., Stoll, J., Mühlbauer, A., Sourdeval, O., Salzmann, M., Heinold, B., and Tegen, I.: Implementation of aerosol-cloud interactions in the regional atmosphere-aerosol model COSMO-MUSCAT(5.0) and evaluation using satellite data, Geoscientific Model Development, 10, 2231–2246, https://doi.org/10.5194/gmd-10-2231-2017, 2017.

Tørseth, K., Aas, W., Breivik, K., Fjæraa, A. M., Fiebig, M., Hjellbrekke, A. G., Lund Myhre, C., Solberg, S., and Yttri, K. E.: Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972 - 2009, Atmospheric Chemistry and Physics, 12, 5447–5481, https://doi.org/10.5194/acp-12-5447-2012, https://www.atmos-chem-phys.net/12/5447/2012/, 2012.

Watson-Parris, D., Schutgens, N., Reddington, C., Pringle, K. J., Liu, D., Allan, J. D., Coe, H., Carslaw, K. S., and Stier, P.: In situ constraints on the vertical distribution of global aerosol, Atmospheric Chemistry and Physics, 19, 11 765–11 790, https://doi.org/10.5194/acp-19-11765-2019, https://www.atmos-chem-phys.net/19/11765/2019/, 2019.

Wex, H., Petters, M., Carrico, C., Hallbauer, E., Massling, A., McMeeking, G., Poulain, L., Wu, Z., Kreidenweis, S., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary aerosol: Part 1 - Evidence from measurements, Atmospheric Chemistry and Physics, 9, 3987–3997, 2009.

Wex, H., McFiggans, G., Henning, S., and Stratmann, F.: Influence of the external mixing state of atmospheric aerosol on derived CCN number concentrations, Geophysical Research Letters, 37, doi: 10.1029/2010GL043 337, 2010.

Wex, H., Dieckmann, K., Roberts, G. C., Conrath, T., Izaguirre, M. A., Hartmann, S., Herenz, P., Schäfer, M., Ditas, F., Schmeissner, T., Henning, S., Wehner, B., Siebert, H., and Stratmann, F.: Aerosol arriving on the Caribbean island of Barbados: physical properties and origin, Atmospheric Chemistry and Physics, 16, 14 107–14 130, https://doi.org/10.5194/acp-16-14107-2016, https://www.atmos-chem-phys.net/16/14107/2016/, 2016.

Wiedensohler, A.: An approximation of the bipolar charge distribution for particles in the submicron size range, Journal of Aerosol Science, 19, 387 – 389, https://doi.org/https://doi.org/10.1016/0021-8502(88)90278-9, http://www.sciencedirect.com/science/article/pii/0021850288902789, 1988.

Wolke, R., Schröder, W., Schrödner, R., and Renner, E.: Influence of grid resolution and meteorological forcing on simulated European air quality: A sensitivity study with the modeling system COSMO-MUSCAT, Atmospheric Environment, 53, 110–130, 2012.

Zängl, G., Reinert, D., Rípodas, P., and Baldauf, M.: The ICON (ICOsahedral Non-hydrostatic) modelling framework of DWD and MPI-M: Description of the non-hydrostatic dynamical core, Quarterly Journal of the Royal Meteorological Society, 141, 563–579, https://doi.org/10.1002/qj.2378, https://rmets.onlinelibrary.wiley.com/doi/abs/10.1002/qj.2378, 2015.