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.

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 (see Hinneburg et al., 2009): "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.

*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.

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.

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.

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: We will add the following sentence in the revised manuscript (p8, l26):
"The ratio between the CCN number and the total particle number as counted by the CPC (condensation nuclei, CN) gives the activated fraction (AF) of the particles. The AF was corrected for multiply charged particle up to three charges by subtracting their apparent fraction from the AF using the charge equilibrium (Wiedensohler, 1988). This multiple charge corrected AF is calculated for each particle diameter and results in a size dependent activation curve for each supersaturation."

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

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

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.

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.