Review of Estimation of Cloud Condensation Nuclei number concentrations and comparisons to in-situ and lidar observations during HOPE experiments by Genz et al.
acp-2019-742

Summary: In this study, the authors use a meteorology model, coupled to an aerosol model that predicts aerosol mass concentrations over the course of several months in Germany where observations are present at two sites within their domain. The aerosol mass concentrations that are simulated in the models are converted to CCN using assumed size distributions, and these CCN concentrations are compared to estimates of CCN from various in situ and remotely-sensed observations in Germany during the same time period. Furthermore, the authors apply scaling factors to the emissions representative of the 1980s, and run the 2013 simulations again with these new emissions to get a sense of what the CCN concentrations were in 1980s for comparison. The authors utilize a wide range of methods to quantify CCN, and a comparison of such methods are useful. However, more details on why these different methods vary would provide a much more compelling study and would improve the interpretation and understanding of not only the 2013 results but also the 1985 results. Furthermore, there are also some critical details that are missing and some missing references. As such, I think this study is of interest to the wider community, but needs additional analyses and more details as discussed below.

Overarching concern

The main focus of this study is on the use of modelled CCN estimates. However, the authors only speculate reasons why their modelled CCN estimates vary from observations (e.g., ammonium nitrate uncertainties, precipitation impacts, particle size distributions, particle composition) and do not provide concrete results explaining why their modelled estimates compare as they do to various observations. All of these speculations could be actually tested in their framework. Understanding the reasons for their 2013 model-observation comparison biases would be especially helpful in assessing the robustness in their 1985 estimates, for which there are no observations and which are based on meteorology from 2013. Ultimately, I am left questioning on how accurate are these 1985 profiles

As such, I think this manuscript can be improved significantly by providing focused analyses on why their model underestimates and overestimates CCN. These include:

1) Assessment of precipitation and surface winds in the model and at their observation site. The authors state that for a 2 day period, the overestimated in the model compared to observations was due to precipitation not being correctly located in their model as compared to observations. Therefore, this potential problem of comparing the model and observations at one fixed site could help explain a lot of overestimation or underestimation of CCN, and if so, then more credence can be given to the various emissions and aerosol assumptions used in this model and post-model processing.

Similarly, airmass trajectories and the advection of aerosol to their specific site could also lead to similar biases and should be tested.

2) The authors state several times throughout the manuscript that there are large uncertainties likely due to the fact that they assumed a fixed PSD for their aerosol size distribution that is required to estimate CCN. It seems reasonable and possible for the the authors to test this assumption, by using a few other PSDs to understand the magnitude of this sensitivity, which would allow for more scientifically rigorous conclusions. This seems computationally feasible as the calculations are run offline. Specifically, the authors note that there PSDs likely have too few particles below 100nm, so at least the authors should run a test with more particles in this part of the PSD.

3) Why does the aerosol vertical profile of CCN change between the 1985 and 2013? There is no clear explanation given.

4) Why was the meteorology from 1985 not taken into account? Is there no meteorology data available from that time period? How can we know that the differences shown in the authors in their simulated 2013 and their simulated 1985 (using meteorology form 2013) would not be different if they actually used meteorology from 1985? The authors even state in their conclusions that the dynamics and thermodynamics (e.g., meteorology) have a large influence on the CCN distribution. Given that the authors treat their 1985 CCN profiles are realistic representations that can be used in future studies and assess the role of emissions reductions between these years, there should be some at least some support, either references or some analysis of the meteorology from these time periods that justify not considering the meteorology.

Specific Comments / Questions

P1, L8: Can the authors extend their CCN values to include a few decimals points, since the units are 10^9 m^{-3}.

P1, L13-14: ‘since chemical composition and the size distribution are less important in these ranges’. This was not shown in this manuscript from my understanding and should be excluded from the abstract.

P1, L16: What does ‘mid-supersaturation regime’ mean? It is suggested the authors use actual numbers here to make it more clear.

P2, L2: It may be a good idea to include the word ‘Europe’ in the first sentence to make this statement accurate and clear.

P2, L4: Should the last sentence of the first paragraph have a reference, possibly the Cherian et al. 2014 reference?

P2, L5-6: Aerosol particles play an important role in the microphysical processes of cloud formation, should have a reference. I believe the proper, original reference for this is: Köhler, 1936.
Köhler, H. (1936), The nucleus in and the growth of hygroscopic droplets, Trans. Faraday Soc., 32(1152), 1152–1161, doi:10.1039/TF9363201152.

P2, L26: The statement says the particle size distribution were calculated using an offline using the Abdul-Razzak and Ghan parameterization. Is this true? I thought the PSDs were assumed, not calculated. Please check.

P2, L31: It is suggested that the authors include some earlier, more original studies of CCN observations. For example, Squires and Twomey, 1966.
Squires, P., and S. Twomey (1966), A Comparison of Cloud Nucleus Measurements over Central North America and the Caribbean Sea, *J. Atmos. Sci.*, *23*(4), 401–404, doi:10.1175/1520-0469(1966)023<0401:acocnm>2.0.co;2.

P2, L32-33: Again there are earlier, more original references that should be considered. For example, Feingold et al. 1998.
Feingold, G., S. Yang, R. M. Hardesty, and W. R. Cotton, 1998: Retrieving cloud condensation nucleus properties from Doppler cloud radar, microwave radiometer, and lidar. J. Atmos. Oceanic Technol.,

P3, L12: The authors state that the activation of aerosol particles depends on their number, which is not true. The activation of aerosol particles only depends on size, composition, and the amount of supersaturation present.

P4, L3: Is there then a discontinuity in the simulation data every 48 hours when the model is re-initialized?

P5, Figure 1. The Julich site is right next to the model domain boundary. If I understand the model set-up correctly, this is a significant concern since if there are winds coming from the west, the aerosol concentrations in the model could be very inaccurate.

P5, L10: The authors state that particles between 50 and 200 nm are most relevant for estimating CCN. This is not strictly true since all aerosol particles can form CCN. Can the authors be accurate with their statements? For example, maybe here the authors mean that particles between 50 and 200nm are most relevant for estimating CCN for supersaturations between some supersaturation range? For shallow clouds with very low supersaturations, the sizes of CCN particles that are relevant may indeed be larger. Similarly, for high supersurations, the relevant particle sizes may be lower.

P6, L10: What does “as written above” refer to? I found this paragraph difficult to follow and suggest the authors revise it to make it more clear.

Table 1: Instead of putting the references in the caption, can the authors put the relevant references next to each species? This would allow a reader who was interested in understanding one of the species assumptions to easily find that without having to look through all of the references listed in the caption.

P11, L30: What causes the significant differences (5-8x different) in OC, SS and DU between the 1985 and 2013 modeled values? Since the emissions do not change, it would be very interesting to understand why these other species change and provide better interpretation of these results.

P12, L7-9: What does ‘large’ and ‘major’ mean? Can the authors be more specific and include numbers?

P12, L18: Why were ammonium sulfate and nitrate underestimated in the model in the first half of spring?

L12, L19: It is unclear what the “factor of 5” is referring to. I am assuming CCN concentrations, but the way it is written it sounds like ammonium nitrate or ammonium sulfate or both?

P13, L1-5: The authors speculate that the overestimation in the modeled CCN may be due to ammonium nitrate uncertainties. However, then they state that actually for a two day period it was due to differences in precipitation and wet deposition. What is the primary cause for the consistent overestimation?

Figure 2c,d: There is very little discussion in the manuscript about the comparisons of the CCN based on the gravimetric observations as compared to the CCNC, which is shown in Figure 2c,d.

P16, L2: “More important” than what?

P16, L12: “observed and measured” -- Are these meaning the same thing?

P18, L4: The authors state that for this analysis a vertical velocity of 1 ms^{-1} was assumed. All the prior analyses had fixed supersaturations, while for this analysis, the authors instead used a fixed vertical velocity. Why did this change?

P18, L4-5: The authors state that “in contrast to the previous analysis, the supersaturation depends on the aerosol composition and varies spatially and temporally.” I think additional statements are needed here or upfront in the manuscript to make this more clear.

P18, L13-14: Why is the difference in shape of the profiles due to differences in aerosol composition (hygroscopicity)? How was this determined, and how did the authors rule out other differences (i.e., differences in aerosol chemistry, differences in meteorology)? There should be much more explanation here.

P18, L15. What do the authors mean when they say the scaling factor represents a mean trend? Are they suggesting there is a linear decrease between the two years? I am not sure the authors can state anything about a mean trend with two points.

P20, L18-19: In their conclusions, the authors state that the thermodynamics and dynamics of the tropopause has a large influence on distribution of aerosol and the vertical profile of CCN. However, the authors state in their manuscript that this was due to aerosol composition hygroscopicity (P18, L13-14). Can the authors clarify what is meant?

P20, L19-20: One of the main results is the differences in the vertical structure of CCN between 1985 and 2013, and the authors state that this vertical structure up to 5km is important for cloud microphysical processes without any references. The two studies that have assessed this are Lebo 2014 and Marinescu et al. 2017 and should be included to support this statement.
Lebo, Z. J. (2014), The Sensitivity of a Numerically Simulated Idealized Squall Line to the Vertical Distribution of Aerosols, *J. Atmos. Sci.*, *71*(12), 4581–4596, doi:10.1175/JAS-D-14-0068.1.
Marinescu, P. J., S. C. van den Heever, S. M. Saleeby, S. M. Kreidenweis, and P. J. DeMott (2017), The Microphysical Roles of Lower-Tropospheric versus Midtropospheric Aerosol Particles in Mature-Stage MCS Precipitation, *J. Atmos. Sci.*, *74*(11), 3657–3678, doi:10.1175/JAS-D-16-0361.1.

Figures 6 and 7. Can the authors make these figures wider, so that readers can more easily see the values that are represented by the lines? Currently, it is very hard to see.