

Interactive comment on “Estimation of Cloud Condensation Nuclei number concentrations and comparison to in-situ and lidar observations during the HOPE experiments” by Christa Genz et al.

Anonymous Referee #1

Received and published: 25 October 2019

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 air-

[Printer-friendly version](#)

[Discussion paper](#)



borne 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.

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 over-estimated because higher condensation/coagulation sinks would prevent the growth of sub-CCN particles into CCN size range.

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

[Printer-friendly version](#)[Discussion paper](#)

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).

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 AS₂₀₁₃ * 5.3 in Table 3?

4. Does “Sulfate” in Table 1 and 3 means “Sulfuric acid”?

5. Table 1: Shouldn’t sigma (μm) in the log-normal distribution be dimensionless?

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

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?

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.

Typo

p.7 L22. Unnecessary comma (Spindler et al., 2013),.

Printer-friendly version

Discussion paper



p.12 L4 repeated “in”

p.14 Line 4. “cf. Figs.6 and ??” What is “??”. Should it be 7?

References

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

Pierce, J. R. and Adams, P. J. (2009). Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates. *Atmos. Chem. Phys.* 9:1339-1356.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-742>, 2019.

Interactive
comment

Printer-friendly version

Discussion paper

