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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inverse modelling of Köhler theory – Part 1: A response surface analysis of CCN spectra with respect to surface-active organic species" *by* S. Lowe et al.

Anonymous Referee #1

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The authors present a new approach to assess the sensitivities of various parameters to CCN numbers. As opposed to numerous previous studies that used a 'one-at-a-time' approach, i.e. varying only one parameter at once, their use of response surfaces can reveal sensitivities over much wider parameter spaces. The focus on surface partitioning and surface tension and it is concluded that careful parameterization of these parameters is needed in order to successfully predict CCN under some conditions, in agreement with previous studies. The current study represents a model framework and the input data are artificially created so that perfect agreement can be achieved. So, therefore it is quite simplistic and does not lead to entirely new results. The sensitivity study for the selected parameters is more comprehensive and the approach might

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be promising to be applied to other problems in the future. However, not all parameters that have been identified as being important for determining the CCN numbers in previous studies have been discussed. Therefore, I think the paper's content and conclusions are somewhat weak and preliminary since only the suitability of the model framework, but not many new results are discussed.

Major comments

1) Mixing state In several previous studies, it has been emphasized that the mixing state of aerosol particles might be one of the most important parameters that determines CCN number in fresh air masses. While it has been addressed briefly in the manuscript, it should be discussed more thoroughly. Could a measure of mixing state be included in the model framework?

2) Previous study on CCN sensitivities In a previous study, Lee et al. (2013) have performed a sensitivity study on a global scale of many parameters using a Monte-Carlo-based approach. This study should be discussed in the light of the results in the current study.

3) Uncertainty in cloud formation The authors state correctly that for data sets other than their calibration data set as used here, a perfect agreement cannot be expected. Some discussion on how accurately CCN numbers should be predicted should be added. For example, all measurements are associated with some measurement error. In addition, other factors influence CCN number (e.g. Lee et al. (2013). Given all uncertainties in the current abilities to predict cloud formation (meteorology, updrafts, emissions, etc), what is the recommendation for a tolerable uncertainty in CCN predictions?

4) Solubility In previous studies, the solubility of organics has been identified as an influential parameter (Riipinen et al., 2015). However, this is not even mentioned in the current paper since all organics are assumed to be completely dissolved. How would consideration of a range of solubilities change the conclusions?

5) Surface-active fraction The discussion of previous studies on the effect of surface tension to CCN activation is somewhat confusing. For example, studies by Noziere et al are discussed that reveal surface tension of $\sim$ 30 nM on aerosols. This number, however, is not relevant for CCN activation unless sufficient material is available to cover the complete particle/'droplet near activation. I suggest discussing the cited references more carefully. What fraction of surface-active material is needed to cause the effects as seen in the current study? Are these fractions realistic?

6) All figures It is not clear to me why the authors do not show all response surfaces (as a supplement). If they choose not to do it, more emphasize should be given why the figures they are showing are shown and not others.

7) Figures Several figures appear blurry and hard to read due to very small font, e.g., Fig. 1 and 3: the indices in the legend are hard to distinguish Fig. 4: Avoid putting the legend box across the lines in the figure

Minor comments

Abstract: The fact that no 'real data' but an artificially created calibration data set has been used, should be mentioned in the abstract.

p. 3, l. 33 (and numerous other places): OF has been defined before

p. 4, l. 18: 'Köhler models' sounds rather colloquial

p. 4, l. 27: Since all the listed studies are model studies, 'observed' should be replaced by 'implied' (or something similar) as the results do not directly refer to observations

p. 5, l. 4: Add a reference for the fact that organic composition has changed since the preindustrial times.

p. 6, l. 11: Sounds odd. 'Is inverse modelling... a well posed problem?' as not the modelling is the problem but the sensitivity to CCN number

p. 11, l. 11: This sentence seems redundant (cf. l. 7)

Technical comments

p. 4, l. 20: supersaturations –> supersaturation

p. 8, l. 8: ‘a’ can be omitted

p. 9, l. 22: SRFA is. . . –> SRFA has. . . (?)

p. 14, l. 8: is –> are

p. 15, l. 17: can assessed –> can be assessed

p. 24, l. 1: in 3 –> in Fig. 3

References

Lee, L. A., Pringle, K. J., Reddington, C. L., Mann, G. W., Stier, P., Spracklen, D. V., Pierce, J. R., and Carslaw, K. S.: The magnitude and causes of uncertainty in global model simulations of cloud condensation nuclei, Atmos. Chem. Phys., 13, 17,8879-8914, 10.5194/acp-13-8879-2013, 2013.

Riipinen, I., Rastak, N., and Pandis, S. N.: Connecting the solubility and CCN activation of complex organic aerosols: a theoretical study using solubility distributions, Atmos. Chem. Phys., 15, 11,6305-6322, 10.5194/acp-15-6305-2015, 2015.

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