

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.

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This study presents a methodology to investigate the sensitivity of CCN spectra to different parameters using different approaches. The results shown are not entirely new and future work is necessary in order to relate them to “real world” characterized by simultaneous measurements of aerosol chemical composition and size distribution, supersaturation, CCN spectra, etc.

The authors would like to take this opportunity to thank the reviewer for their constructive comments; we believe that they have added to the quality of the manuscript as a whole. The purpose of this study was to present the development of a new framework for probing sensitivity of aerosol activation to pro-

cesses which have been studied in isolation over a number of years. By embracing an inverse modelling approach to aerosol-CCN closure, we not only build a framework for sensitivity analysis, but also a method of diagnosing both structural and parametric uncertainties in model CCN predictions by simultaneously matching input parameters and model output. In section 5.3 of the revised manuscript, additional material is presented to highlight the importance of calibration data resolution and natural variability in CCN measurements as a natural first step towards future work with observational data. Our response is presented in bold text following the reviewer's comments. Any referencing of sections, pages or line numbers given in the response pertain to those of the revised manuscript.

Major comments:

1. The theoretical investigations shown here were performed at a fixed temperature, using literature data for three aerosol types: marine, polluted continental and rural. The organic aerosol and surface tension were assumed to be the same for all aerosol types. The possible effects of this data are not discussed in the manuscript.

Preliminary testing of temperature was performed and negligible sensitivity was found. We therefore chose to exclude it from our analysis. The reviewer raises a good point with regard to the organic aerosol data. Unfortunately, as the organic aerosol fraction exhibits such high levels of complexity and spatial and temporal variability, in order to be more precise than we have been is difficult. By providing generous ranges for our model organic we hope to encompass all possibilities that may be realised in the real atmosphere. Indeed, once coupled with the MCMC algorithm (part 2) the framework developed in the present study provides a useful tool to constrain such parametric uncertainties by exploring all possibilities in the complete parameter space.

2. The manuscript looks like a report, most of the section 4.2 may be moved in a Sup-

plementary Material. The results shown in it have to be presented in a more synthetic and comparative manner. It is not evident how much the sensitivity to parameters depends on the approach used in a quantitative manner. This is also not clear in Table 3 which is not actually discussed. Also, it has to be moved from Conclusions to Results.

We agree that the manuscript could be more concise, therefore the text has been substantially reduced and formulated in a more synthetic and concise manner. A supplementary document that contains all response surface analysis has now been attached with the manuscript. However, as inverse modelling of CCN spectra has not been performed before, we believe that the response surface analysis contained in section 5.2 (previously 4.2) is essential to the understanding of how to correctly couple the framework to automatic search algorithms, such as MCMC, which will form the focus of a part 2 study. Therefore we choose to keep the section in the main text.

3. Such a study may be very interesting if it succeeds in showing the limits/differences due to the different approaches used in modelling aerosol activation and quantify the acceptable/relevant uncertainties of measured parameters of fundamental importance in these approaches. The latter information may be important for planning future field campaigns and for development of instruments.

The reviewer raises a very good point here and we have chosen to add additional material to section 5.3 to address the issue at hand. Therein we have discussed at length the importance of information content for various definitions of the calibration that may arise due to different instrumentation. We also account for the natural variability in these considerations. It was concluded that, when accounting for natural variability in the analysis, it would be challenging to correctly minimise the OF based on the information content of a calibration data set measured by a typical CCN counter. However, correct minimisation of the OF was still achieved when corrupting the high resolution calibration data set with randomly generated natural variability. As such, this result should serve

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as a recommendation for the development of instrumentation for high resolution measurements of CCN spectra in-situ.

Minor comments:

The term “posedness” is not commonly used, replace or explain it better

The term posedness has been replaced throughout the manuscript with ‘well-posed’, ‘ill-posed’ or similar.

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