Response to anonymous reviewer 2: Partridge et al., 2011.

The authors thank anonymous reviewer 2 for insightful comments on the manuscript. The reviewer provided several suggestions for improving the readability and quality of the manuscript. We have followed the suggestions in most cases, and our detailed response is outlined below.

Summary of main changes:

- Clarified the application of the study in both the introduction and goals sections.
- Introduced response surfaces as a means to study susceptibility of a CDNC distribution to perturbations in aerosol physiochemical parameters as well as the updraft velocity for four distinctly different aerosol environments.
- Compressed all sub-sections of section 3 so that the main points are conveyed clearer by summarizing our main findings relevant for both modelers and instrumentalists.

Response to general comments:

G1: It is not fully clear for what purpose this article is written. In general, model sensitivity analysis should be the first step in any modelling application. In that sense, the text would make a good material for a textbook on aerosol modelling. Almost every modeling exercise in any applied field contains numerous iterations and reformulations. Many times by trial and error, sometimes by using some well established methodological recipes. It is not necessary to report these steps towards the final model formulation unless the methods used are of interest by novelty, for example. The lack of any real life observations makes the exposition to be more of a pedagogic nature, also.

RG1: The paper series is designed to investigate cloud-aerosol interactions for both synthetically generated observations and real life observations by coupling automatic search algorithms to a pseudo-adiabatic cloud parcel model.

Part A covers applying (to the author's knowledge) for the first time an automatic search algorithm to a pseudo-adiabatic cloud parcel model, and response surfaces (cf. further discussion below) as a means to visualize cloud-aerosol interactions with respect to CDNC distribution susceptibility and the posedness of the cloud-aerosol inverse problem.

Part B covers applying (to the author's knowledge) for the first time MCMC methods for inference of the posterior probability density function of the parameters to study cloud-aerosol interactions. By first obtaining parameter sensitivity from synthetically generated observations (part B) we will be able to compare the results from this method to previous synthetic studies, and also to when the sensitivity is conditioned on real droplet size distribution observations.

In a following study we will apply MCMC simulations using real world observations from the MASE II campaign (Partridge et al., 2011; manuscript in preparation).

With paper A, we would like to emphasize the importance of first using synthetically generated measurements with inverse modeling of cloud-aerosol interactions to benchmark the analysis and results. As we have shown there are numerous facets to the problem (chaotic response surfaces related to an interpolation in clean environments; multiple local minima; non-identifiable parameters that require fixing to their true values) and these all need considering before coupling a cloud parcel model to an automatic search algorithm.

According to the suggestion of the reviewer, the layout of the paper series and the purpose of paper A is now defined/explained clearer in section 1.2. In addition we provide an application of response surfaces to investigate CDNC susceptibility to four distinctly different aerosol environments, the results for which are provided in section 3.4.

G2: The authors emphasize the choice of the objective function (OF). In general, OF includes the parameterization of the unknowns (e.g. the transformations used) the choice of parameters to estimate, the observations (i.e. the design of the measurement processes), the forward model formulation, and also the statistical distribution of the error term and the prior distributions for the parameters. Again, the formulation of all these is important in any inverse modelling. To my mind, the authors do not clearly distinguish between these different elements. Everything seems to be studied only in connection with the difficulties there will be in using a numerical optimization algorithm. The questions that are more interesting would be of the following type. What observation can and should be collected? What is the information content of these measurements? How should the model be parameterized with respected to the unknowns. What kind of prior information is available and needed to make the unknowns identifiable? Are we aiming to design a new measuring instruments? Are we trying use existing instruments and to perform experiments with maximum amount of information about a given model?

RG2: The reviewer raises an important point. The implications for both instrumentalists and modelers have been made clearer by summarizing the most important points at the end of sections 3.1 and 3.2. Also the focus of the paper has been shifted towards investigating the susceptibility of CDNC distributions. In order to highlight the application of response surfaces we have now included susceptibility analysis “application” in the manuscript so that the sensitivity to four distinctly different aerosol environments is presented (section 3.4). To achieve this we have modified the definition of the calibration data (section 2.2.2-2.3) as the focus of the paper is now to present susceptibility as well as the parameters that will need fixing prior to coupling the a cloud parcel model with an automatic search algorithms, and the limitation of the moving centre framework and associated interpolation for clean clouds for the successful solution to the inverse problem.

G3: The authors give a hint of a more comprehensive modelling approach in a forthcoming "part 2" of the paper. For studying parameter uncertainty and identifiability in non-linear models the methods offered by Bayesian statistical analysis and MCMC simulation algorithms are superior in many ways to standard classical sensitivity analyses. Using MCMC method, the multi-dimensional cost function surface (the posterior distribution of the model unknowns) can be explored fully. Non-identifiability can be diagnosed by MCMC, and by using suitable efficient and adaptive MCMC methods, it is usually possible to gather information even if some of the parameters have very wide marginal posterior *distributions. The Bayesian approach allows for "fixing" the non identified parameters by a prior distribution instead of fixing them to a single value. Instead of two dimensional surfaces that are conditioned on some given values of all the other unknowns, one or two (or higher if needed) dimensional marginal posterior densities can be easily constructed from the MCMC chain.*

RG3: The purpose of part A and B in the paper series are now more clearly explained in section 1.2. The point raised by the reviewer regarding using MCMC to study parameter uncertainty the reviewer is very interesting and will be discussed/analyzed in part B of the paper series., To summarize, yes, we can diagnose insensitive parameters with MCMC; the size of the marginal distribution is a measure of sensitivity, as discussed and showed in our subsequent papers. Such MCMC approach will take into consideration all parameters simultaneously, whereas response surfaces considered herein only consider two parameters. Yet, if the parameter sensitivity is poor in two-dimensional plots by varying only two parameters, it is expected to be even worse if all parameters are varied. The compensation effects are even bigger then, and thus the sensitivity will only be worse. Aside from this response surface analysis provides a clearer way to understand other problems associated with the inverse problem, for instance those associated with the necessary post-processing to a fixed size grid, as we demonstrate with the interpolation. Thus, we hope this paper will facilitate more efficient research in this field by allowing future studies to avoid these pitfalls.

G4: Without seeing the "part 2" of the article, I would suspect that some actually interesting geophysical results could be acquired by the MCMC analyses. So, to conclude, I'm looking forward for the part 2 that would describe full statistical uncertainty analysis by MCMC or related methods. Also, applying the methods to real observations would make the methodology more interesting for the typical readers of ACP. My suggestion is to write the planned "part 2" using statistical inversion methodology and with real data in addition to simulated data. Consider including the most important lessons learnt from the sensitivity analyses of "part 1" as one section in the new article.

RG4: The purpose of part A is to introduce the applicability of inverse modelling to cloud-aerosol interactions, and response surfaces as a method to investigate both the posedness of the problem and parameter sensitivity. We have now modified the results of the paper, shifting the focus towards providing a CDNC susceptibility analysis for four distinctly different aerosol environments. We believe that this method provides a visually informative way to investigate cloud-aerosol interactions across 2 dimensions and complements part B in which we infer the sensitivity to all parameters simultaneously. It is clearly showed from our response surface analysis that updraft velocity is a crucial parameter to accurately measure, and that the CDNC is very susceptible to changes in aerosol physiochemical conditions for clean aerosol environments.

Response to major comments:

M1: In statistics the term "response surface" has been traditionally used for analyses where one is interested in the optimal value of some process model response and a set of designed experiments is used to form an approximate (usually quadratic or surrogate) model near the optimum response. These analyses are usually graphically aided with two dimensional plots of the "response surfaces". Here the term "response" is used for the value of the objective function. I must admit, that by looking at the title of the article, I was expecting to find a different kind of response surface analysis. These plots used in the article could simply be called parameter sensitivity plots or conditional log likelihood surfaces, for example.

RM1: We think this may be a misunderstanding; we do not call the plots conditional log-likelihood surfaces as we do not perform any Bayesian analysis in this study, nor do we use a likelihood function. That is for the next paper (part B). In this study we do not use a log-likelihood function; although we could from the RMSE. Thus in this study we use the terminology response surfaces as that is the name in the literature (Sorooshian and Arfi, 1982; Toorman et al., 1992; Šimůnek et al., 1998 and Vrugt et al., 2001).

M2: *It is not explicitly stated, whether exactly the same forward model is used both to simulate the synthetic data and to perform the inversion. Even if only studying or developing an inverse method it is advisable to generate the data with more a comprehensive model (with a higher resolution, for example), to avoid so called inverse crime [Kaipio & Somersalo, 2004].*

RM2: In this study we use the same pseudo-adiabatic cloud parcel model to generate synthetic data and to perform the inversion analysis. This is (now) clearly stated in section 2.1.

We do not agree with the reviewer that it is advisable for the purpose of this study to generate the synthetic observations with a more comprehensive model. If this was done then we would not only have parameter uncertainty but also model uncertainty. This would corrupt the response surfaces tremendously, and would make it very difficult to explain the results. The goal of this paper is to show the various elements of inverse modeling with a specific focus on cloud-aerosol interactions. If we follow this idea of using a more complex model to create out data, we cannot longer benchmark our results. We would not know which parameter values are the "true parameters" for the simplified model if we use another model to generate the calibration data.

Despite this, the comment of the reviewer is useful, and we believe it should be investigated in future work. This would answer the important question of how model uncertainty affects the optimized model parameters and their nonlinear uncertainty. Yet, this is beyond the scope of the current paper that attempts to demonstrate the usefulness and applicability of inverse modeling in modeling cloud-aerosol dynamics. The next step, (also after the subsequent paper) would be to provide a formal treatment of different types of error, including model structural inadequacies. This subject is very difficult and currently hotly debated in different fields.

M3: *The main message seems to be that the inverse problem presented is difficult to solve, and care must be taken when defining the objective function, choosing the optimization method and selection the measurements. The information content studies show that: "the inclusion of the right type of information, not necessarily more information that is most important". Not very strong or helpful result to offer for anyone trying to solve similar problems.*

RM3: We agree that we do not provide an immediate solution to the information content problem, yet this paper is just to show that the measured size distribution contains relatively little information, and that this limits how many parameters can be estimated. We now highlight this in a clearer way by rewriting section 3 and summarizing our main findings at the end of sections 3.1.3.2. We demonstrate how the right type of information is more important than more information with respect to the lognormal aerosol parameters describing the smaller Aitken mode. It would be very difficult to calibrate these parameters using any form of the droplet size distribution alone, and therefore, they require the inclusion of the interstitial aerosol in the calibration data set. Measuring the interstitial aerosol in clouds will also help benchmark cloud models to a higher degree of accuracy, especially for polluted conditions. We also show that for polluted clouds it would be very useful in future measurement campaigns to measure the interstitial aerosol. This will help constrain the inverse problem and allow a more rigorous benchmarking of simple cloud-parcel models to help improve parameterization of droplet activation. Other possible ways forward is to use multiple different height levels in the objective function or to perform a wavelet post-processing on the calibration data instead of an interpolation yet this is beyond the current study.

M4: *A simple Monte Carlo analysis is compared to one specific deterministic optimization method. Again, the conclusion is not very surprising. "A brute force MC approach is not only inefficient, but potentially also misleading", but optimization method can find "true" optimum that was used in generating of the synthetic data.*

RM4: In this first application of an automatic search algorithm to a pseudo-adiabatic cloud parcel model (to the authors knowledge) we are just trying to show that MC can lead to misleading results, and that optimization is required to find the optimum solution. This links Part A to Part B very clearly. MC is still widely used in the community, including publications in ACP so it is useful to portray for inverse modelling novices the advantages of embracing sophisticated search algorithms.

References:

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