

## Response to anonymous reviewer 1: Partridge et al., 2011.

The authors thank anonymous reviewer 1 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: Introduction. In the last section, the authors will give a hint regarding the potential applications of the method. Please make this more concrete – what are exactly the applications (what measurements are needed and what aerosol properties can be inferred)? Why the manuscript is named as “Towards...”? What will the other parts of the series contain?*

R1A: We have made the potential applications of the method more concrete in the introduction by introducing a susceptibility analysis of the aerosol physiochemical and meteorological parameters in the study as an application for four distinctly different aerosol environments. This is stated now at the end of the introduction with some rewording of the other text to highlight the advantages of response surface analysis to identify what measurements are needed for different aerosol environments. In addition we have reworded the goals (section 1.2) of the manuscript to accommodate the new application provided by our response surface analysis.

R1B: The manuscript was initially titled “Towards” since we saw this study as an introduction to cloud-aerosol inverse modeling, outlining a methodology that will aid in the guiding of future studies away from potential pitfalls using a full MCMC analysis with cloud-parcel models. As we have now changed the focus of the paper and included an application in this manuscript the “Towards” has now been removed from the title.

Many studies have probed cloud-aerosol interactions using synthetically generated observations (Chuang et al., 1997; Nenes et al., 2001, 2002; Feingold et al., 2003). In part B we aim to further these studies by performing a full MCMC analysis of the cloud-aerosol inverse problem also using

synthetically generated cloud droplet size distributions. We would like to highlight that it is important to first use synthetically generated measurements with inverse modeling of cloud-aerosol interactions so that we can benchmark our analysis and results, as to our knowledge an automatic search algorithm has never been applied to this specific scientific question before.

To facilitate part B we demonstrate in part A why the cloud-aerosol inverse problem will be particularly difficult to solve. This insight as to the problems associated with the cloud-aerosol inverse problem will aid future improvements to inverse modelling studies using cloud parcel models with synthetically generated observations for which it is useful to investigate as many parameters simultaneously as possible. To satisfy the reviewers concerns regarding the potential application of response surfaces we now also in part A present a separate application using response surfaces as a tool to present in a visual and transparent manner 2D parameter sensitivity for four aerosol environments. The results from this analysis are presented in section 3.3. This provides a natural step to part B in which we derive the sensitivity towards all parameters simultaneously.

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). By first obtaining parameter sensitivity from synthetically generated observations (part B) we will then be able to compare these results to when the sensitivity is conditioned on real droplet size distribution observations.

*G2: Method validation. The authors base their approach on using an adiabatic air parcel model. This also implies that the method is only applicable to measurements conducted under adiabatic conditions. This point should be mentioned. Also, a discussion regarding how to verify this (i.e. which measurements are needed to ascertain adiabaticity) should be included.*

R2: The reviewer brings up an important point. Adiabatic cloud parcel models have frequently, and successfully, been used as inter-comparison tools with field measurements to estimate the impact of aerosol size/composition for liquid clouds (e.g. Hsieh et al., 2009). It is recognised however that few clouds are adiabatic in nature (especially far away from cloud base) This is not discussed within this paper as it is covered in more detail in a following study when we use real world observations from MASE II. In this study (Partridge et al., 2011; manuscript in preparation) we use the available liquid water content (LWC) data to filter for different adiabatic conditions as has been performed in previous studies (Lu et al., 2009).

*G3: Choice of the objective function. Please provide an explicit equation for the chosen objective function. Now it is extremely hard to understand the discussion in the last two paragraphs of section 2.3. The same applies to the exact definition of the objective function. Please re-write the section to make it more accessible. In particular, it is hard to understand how two different quantities are included in the objective function. Also, because of the moving sectional approach, the diameters at a certain bin  $i$  do not match when comparing the model and measurements/synthetic data. How did the authors overcome this problem? This is crucial to the manuscript so clarify this point when discussing about “X and Y components of the size distribution” (the term itself is rather vague).*

*A second issue which I could not grasp is how the authors could have problems when interpolating the data to a grid with a different resolution. In common modeling applications, adiabatic air parcel models are run with much more number of bins compared to the resolution of the applied instruments. This should make the interpolation rather trivial without causing any dents/spikes to the re-mapped data. Also, have the authors tested different interpolation algorithms found in the literature? To me, this seems more like a technical problem rather than a fundamental one.*

R3A: In the present version of the manuscript it was necessary to modify our chosen objective function so that it is no longer split into its X and Y components in order to provide a stronger application of response surfaces in this study rather than only focusing on the difficulty of the inverse problem. Thus the calibration data is now defined as the  $dN/d\log dp$  size distribution function interpolated to a fixed size grid. We have provided an explicit equation for this objective function in section 2.3 (Eq. 2). Section 2.3 has also been rewritten to make the section clearer.

The previous definition of the calibration data used in the objective function (Eq. 2 in previous manuscript) in which we split the droplet size distribution into its X and Y components as an alternative method has been moved to the end of section 3.3. This method is in essence an algorithmic trick; we include the radii value; because the different simulations will have different radii values; the differences with the true ones is then used as a distance, and helps the algorithm go in the right direction. The reviewer questions:

R3B: The interpolation presents a problem for applying inverse modelling methods to very clean clouds. Regardless of the interpolation scheme used the same problem will arise:

If you have two different parameter combinations that are both completely off in terms of radii values compared to the original size distribution; then both will have an interpolated value of the size distribution of 0. This will result in the same objective function, and the algorithm does not know how to improve the estimates; it will become stuck in an area with zero values, severely hindering convergence towards the true optimal solution.

Therefore it is important that this is highlighted to facilitate future studies on inverse modelling of cloud–aerosol interactions using synthetically generated data from a cloud parcel model. To make this point clearer in the manuscript we extended the discussion at the end of section 3.1.

*G4: Section 3. As mentioned above, please compress all sub-sections so that the main points are conveyed. Also, this should save some space to include a discussion about atmospheric applications – what measurements (including aerosol related) are needed to constrain what parameters? The authors need to address this question as because it is known already from forward-modeling that different parameter combinations yield the same cloud droplet or CCN number concentrations. Related to this, can the authors identify parameters that cannot be constrained using the information that the current aerosol instrumentation provides?*

R4: Following the reviewers suggestion we have compressed all sub-sections so that the main points are conveyed. 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 with specific relation to insight significant for both instrumentalists and modelers.

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