



A study of marine stratocumulus clouds using an inverse modelling approach

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The difficulty in untangling relationships among aerosols, clouds and precipitation has been partly attributed to the inadequacy of existing tools and methodologies. Current parameterisations used to estimate cloud microphysical properties are usually based upon some form of parcel model theory. These parameterisations are typically validated with closure studies; however, few have employed detailed statistical methods to assess model sensitivity and compare simulated and measured cloud droplet spectrum (aerosol-cloud droplet spectrum closure). One approach is to embrace a Bayesian inverse modelling framework and invoke posterior probability density functions of model input parameters by coupling an adiabatic cloud parcel model to a Markov Chain Monte Carlo (MCMC) algorithm.

This study presents a Bayesian inverse modelling approach to simultaneously assess the ability of an adiabatic cloud parcel model to match in-situ measurements of the droplet size distribution in a cloud as well as model parameters describing the updraft and different aerosol microphysical properties (herein termed calibration parameters). Our methodology is tested using observations from two clean (average accumulation mode number concentration $< 60 \text{ cm}^{-3}$) and two polluted clouds (average accumulation mode number concentration $> 100 \text{ cm}^{-3}$) observed during the Marine Stratus/Stratocumulus Experiment (MASE II) campaign. The framework capitalises on recent developments in MCMC simulation and retrieves the most likely parameter values as well as a sample set of their respective underlying (posterior probability density function) uncertainty. This distribution provides the necessary information to efficiently and in a statistically robust manner assess both the global sensitivity of aerosol physiochemical and meteorological parameters, and the suitability of cloud parcel models for describing the evolution of cloud droplet size distributions in marine stratocumulus clouds.

We demonstrate that the updraft velocity is the most important calibration parameter for describing the observed droplet distribution for each cloud case, corroborating previous findings. The accumulation mode number, shape and size are found to be generally more important than chemistry for the cloud cases investigated.

Overall, the MCMC algorithm successfully matches the observed droplet size distribution for the majority of the cloud cases investigated. In doing so, however, the subsequent agreement between certain derived and measured calibration parameters is poorer. An important result from this analysis is that for certain calibration parameters, consistent patterns of deviation were found in the posterior distributions for all the clouds included in this study. This finding indicates that either there is systematic sampling or averaging artefacts in our observations, or our adiabatic cloud parcel model omits or consistently misrepresents processes and/or parameter(s) required to accurately simulate the observed droplet size distributions. By repeating our inverse methodology with more calibration parameters of which current measurements are uncertain (surface tension, mass accommodation coefficient), we find that it is likely that the process description within the current formulation of the pseudo-adiabatic cloud model used in this study misses a dynamical process rather than parameter(s).