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Inverse modelling of Köhler theory - Part 1: A response surface analysis of CCN spectra with respect to surface-active organic species

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In this study an inverse modelling framework for the calculation of CCN spectra is developed to facilitate a more robust treatment of evaluation of Köhler models against observations.

To achieve this, we define an objective function that provides a diagnostic metric of the deviation of modelled CCN spectra from observations as a function of input parameters. This allows for the assessment of model accuracy while simultaneously examining global parameter sensitivities and identifying parameter interactions across all atmospherically relevant supersaturations, corresponding to a broad range of cloud types and updraft velocities.

The focus of this study is two-fold. Firstly, we assess the feasibility of inverse modelling as a new methodology for aerosol-CCN spectra closure. To achieve this goal, responses in the objective function to parameter perturbations in 2D cross-sections of the complete parameter space, response surfaces, are used to examine the likelihood of our chosen objective function containing enough information to constrain the model input parameters considered using automatic search algorithms. Secondly, these response surfaces are employed to conduct an extensive parametric sensitivity analysis and subsequently rank the relative importance of aerosol physiochemical parameters in determining CCN spectra. Using Köhler theory to model CCN concentrations requires knowledge of many physiochemical parameters, some of which are difficult to measure in-situ at the scale of interest. Therefore, novel methodologies, such as the one developed here, are required to probe the entire parameter space of aerosol-cloud interaction problems and provide global sensitivity analyses to constrain parametric uncertainties.

Partitioning of surface-active species from the bulk to the surface phase can alter the point of CCN activation. Therefore, the analysis conducted here is carried out for a standard Köhler model as well as more complex Köhler models accounting for the partitioning process.

The response surface sensitivity analysis identifies the accumulation mode concentration and surface tension to be the most sensitive parameters. The organic:inorganic mass ratio, insoluble fraction , solution ideality and mean diameter and geometric standard deviation of the accumulation mode showed significant sensitivity while chemical properties of the organic exhibited little sensitivity within parametric uncertainties. Parameters such as surface tension and solution ideality, can introduce considerable parametric uncertainty to models and are therefore particularly good candidates for further parameter calibration studies.

A complete treatment of bulk-surface partitioning is found to model CCN spectra similar to those calculated using classical Köhler Theory with the surface tension of a pure water drop, as found in traditional sensitivity analysis studies. In addition, the sensitivity of CCN spectra to perturbations in the partitioning parameters K and Γ was found to be negligible. As a result, this study supports previously held recommendations that complex surfactant effects might be neglected and continued use of classical Köhler Theory in GCMs is recommended to avoid additional computational burden.