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A simple method for including cooling rate dependence in a deterministic ice nucleation model

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The formation of ice in the atmosphere can extensively alter cloud properties and have a significant radiative impact on the Earth's energy budget. In nature ice nucleation within supercooled droplets is a fundamentally stochastic process, but it is often assumed that the stochastic nature of ice nucleation is second order to the particle-to-particle variability of ice nuclei (IN). Neglecting the time dependence has led to the deterministic approximation, commonly called the singular model. It is known that aerosol particles possess a wide range of ice nucleating abilities, which arises through the presence of multiple components. Each component can be characterised through its activity and time dependent nature. Laboratory studies on atmospherically relevant aerosol components show that ice nucleation by some are measurably time dependent. At low cooling rates this could potentially lead to an under predicted ice production rate when following the deterministic approach. It is therefore apparent that the characterisation of atmospheric IN is required along with a modelling framework that predicts ice formation in a simple, but physically realistic way.

In the first part of this study we use the Met Office KiD model to assess the importance of time dependence over a range of cooling rates. We use parameterisations based on recent laboratory data in which the time dependence of immersion mode nucleation was quantified. We show that even a small discrepancy in the ice production rate can significantly affect the evolution of the cloud hydrometeor species. In the second part of this study the Multiple Component Stochastic Model (MCSM) is used in an idealised model to examine how the distribution of nucleation sites and the temperature dependence of the nucleation rate ($\mathrm{dln}J/\mathrm{d}T$) affects the time dependent nature of an IN. We find that, regardless of the site distribution and mean activity, it is solely the temperature dependence ($\mathrm{dln}J/\mathrm{d}T$) that controls the time dependent behaviour. Finally we show that this simple relationship can be used to determine the change in nucleation rate for a change in cooling rate. This has been applied to the deterministic approximation to provide a computationally efficient freezing model that correctly represents both the distribution of nucleating abilities and time dependent nature of atmospheric IN. Using recent laboratory data of immersion mode nucleation on atmospherically relevant IN (including kaolinite, feldspar and soot) this cooling rate dependent deterministic approximation is shown to be able to reproduce the ice production rates for a range of IN species over a range of cooling rates. In the future we recommend that the cooling rate dependence of ice nucleation is reported along with laboratory or field based deterministic parameterisations in order to capture both the singular and stochastic nature of ice nucleation