Time and Temperature Dependence of Freezing Nucleation in a Cloud Parcel Model

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Abstract. Most models of primary ice formation in clouds describe the number of immersion freezing events as dependent either on time or on temperature. This paper reintroduces a model that bridges these viewpoints in an empirical basis. This is important for modeling ice evolution in cloud parcels whose temperature history is not restricted to steady cooling but involves evolution at constant temperature. Stratocumulus and altocumulus are two examples of specific relevance.

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INTRODUCTION

We reformulate the time-dependent freezing rate model (TDFR; [4]), combining temperature- and time-dependence, and elucidate how immersion-freezing experiments can be utilized to constrain the model. The utility of this approach is apparent in its application to the initial appearance of ice particles within clouds. An example is presented after the development of the TDFR.

TEMPERATURE DEPENDENCE

We consider an ensemble of many liquid drops undergoing steady cooling followed by a period with the temperature remaining constant. We will express the number of ice nucleation events as the sum of those corresponding to the steady cooling and constant-temperature segments. This sequence is applicable to cloud parcels as well as to laboratory experiments. Use is made of the freezing rate defined in terms of the number of drops freezing per unit time

$$R(T,t) = \frac{1}{n_u(T,t)} \cdot \frac{\Delta n_u(T,t)}{\Delta t} \qquad (1)$$

where n_u is the number of unfrozen drops and Δt is an interval of time corresponding to a count of freezing events (Δn_u) [4].

The relevant characteristic of the ensemble is the number of ice nuclei per unit volume of liquid as a function of temperature $K(T)$; a quantity which can be determined in laboratory experiments or can be predicted based on some aerosol to cloud transfer model. In [5] this quantity is defined as the cumulative nucleus

spectrum. The relationship between this quantity and cumulative number of ice crystals per unit volume of air is

$$N_{ice}(T) = \frac{LWC}{\rho_w} \cdot K(T) \tag{2}$$

where the scarcity of nuclei is assumed to ensure that the average concentration of nuclei per drop is less than unity. $K(T)$ is most conveniently determined by evaluating $R(T,t)$ during steady cooling. The relationship between the freezing rate and $K(T)$ for cooling at a steady rate $r = dT/dt$ is

$$K(T) = \frac{1}{|r| \cdot V} \cdot \int_0^T R(T') \cdot dT' . \tag{3}$$

It is worth emphasizing that V in this relationship is the volume of a laboratory sample unit, corresponding to the determination of $K(T)$, or the volume of a single member of a cloud drop ensemble. Dilution or evaporation of the drops will change $R(T')$ but only in minor ways, for example due to changes in soluble ion concentrations and other similar effects.

TIME DEPENDENCE

As shown in [4], two corrections are needed to the foregoing in order to account for time-dependence. First, the rate of cooling, related to updraft velocity, can be accounted for with an offset in temperature, depending on $|r|$.

$$N_{ice}^{cc}(T) = \frac{LWC}{\rho_w} \cdot K(T + \alpha) . \tag{4}$$

The second step is to determine the freezing rate when temperature reaches a steady value T_S. The key finding for this is that the rate decreases exponentially with time from an initial value that is a fraction of that just prior to cessation of cooling

$$\frac{R^{ct}}{R^{cc}} = \frac{Constant\ Temperature\ Freezing\ Rate}{Constant\ Cooling\ Freezing\ Rate} = \omega \cdot e^{-\beta \cdot (t - t_S)} \tag{5}$$

with t_S as the time at the start of a constant-temperature segment and with ω and β as parameters [4]. From a limited number of experiments, Vali ([4]) reported $\omega \approx 0.46$ and $\beta \approx 0.23$ min^{-1}; these parameters need yet to be evaluated for a range of conditions.

From the foregoing it follows that the cumulative number of ice crystals, per unit volume of air for the constant-temperature period can be expressed as

$$N_{ice}^{ct} = \frac{LWC}{\rho_w} \cdot |r| \cdot \left[\frac{N(T_S) - N(T_S + \Delta T)}{\Delta T} \right] \cdot \int_{t_S}^{t} \omega \cdot e^{-\beta \cdot (t' - t_S)} dt' . \tag{6}$$

The term in square brackets expresses the cumulative number of nuclei, per unit volume of liquid, active between $t_S - \Delta t$ and t_S (temperature between $T_S + |r| \cdot \Delta t$ and

T_S). This quantity is defined in [5] as the differential nucleus spectrum evaluated at T_S; it is designated as $k(T_S)$. With this substitution Eq. 6 becomes:

$$N_{ice}^{ct} = \frac{LWC}{\rho_w} \cdot |r| \cdot k(T_S) \cdot \int_{t_S}^{t} \omega \cdot e^{-\beta \cdot (t' - t_S)} dt' \tag{7}$$

The count of ice crystals, specific to a unit volume of air, is evaluated as the sum over both the cooling and steady temperature periods:

$$N_{ice}^{ct} = \frac{LWC}{\rho_w} \cdot \left(K(T + \alpha) + |r| \cdot k(T_S) \cdot \int_{t_S}^{t} \omega \cdot e^{-\beta \cdot (t' - t_S)} dt' \right) \tag{8}$$

RESULTS

Eq. 8 is the TDFR model for the air parcel history here considered. It incorporates both temperature and time into a description of the ice crystal concentration. Inputs to this equation are the differential and cumulative forms of the nucleus spectrum and the parameters α, β and ω. As mentioned earlier, the spectrum can be determined empirically or predicted from an aerosol to cloud water model. The parameters α, β and ω can be determined empirically but since they are manifestations of the basic kinetic nature of ice nucleation, in principle they can be derived from theory. It may be noted that there is no specification of the count of cloud drops, within the air parcel, due to the assumption that the count of ice particles is much smaller. It should also be noted that the groupings $LWC \cdot K(T)/\rho_w$ and $LWC \cdot k(T)/\rho_w$ represent the cumulative and differential spectra of ice nuclei that have entered into the cloud water and that these will be constant, in the absence of aerosol scavenging, or will increase because of it.

The TDFR model is exercised in **Figures 1a-1b** with a cumulative nucleus spectrum from an analysis of rain collected in Colorado ([6]). Assumed cloud properties (temperature lapse rate and updraft), are representative of a high-based summertime cumulus. The appearance of ice crystals during the cooling (updraft) portion of the simulation, with r = -1.2 ^{o}C/min (black lines), is based on Eq. 4. The gray line segments represent nucleation occurring at constant temperatures T_S =-6 and -11 ^{o}C. The ice crystal concentration at constant-temperature is based on Eq. 7 with time-dependent parameters from [4]. These simulations demonstrate that the quantity of ice produced at constant temperature is comparable in magnitude to that produced during cooling.

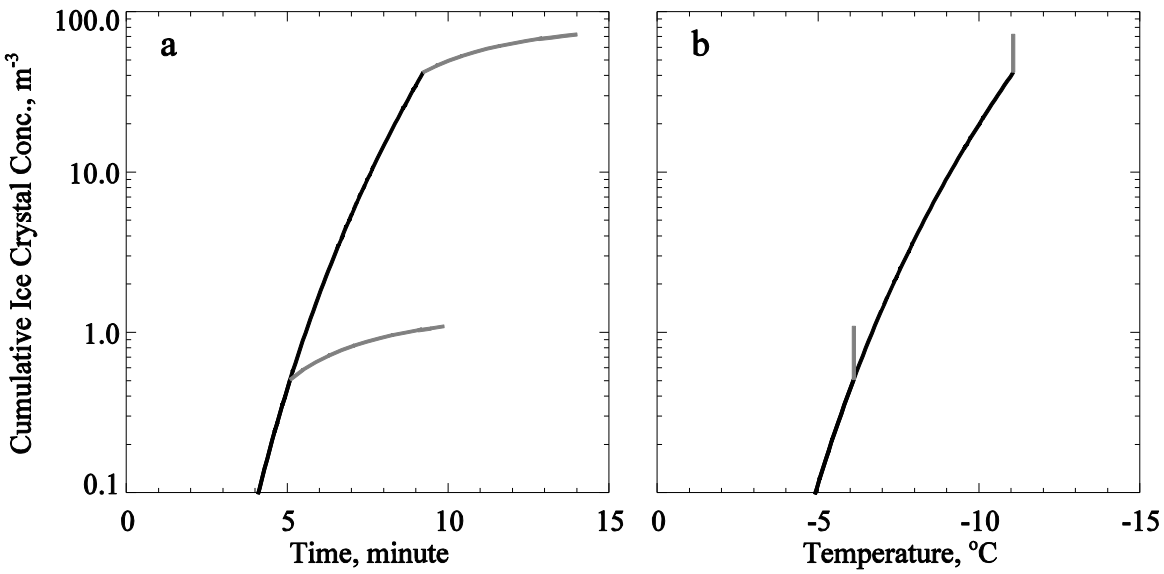

FIGURE 1 – Predictions of the TDFR model for cloud scenarios with constant temperature at T_S = -6 and T= -11 °C (gray lines) following constant cooling at r = -1.2 °C/min (black lines). Parameters describing the time-dependent ice crystal concentration are from [4]. In both scenarios the cloud properties are: cloud base temperature = -1 °C, temperature lapse rate = 5 °C/km and updraft = 4 m/s. The cumulative spectrum is for summer rain in Colorado ([6]).

CONCLUSION

Laboratory results were used to model the evolution of the number of ice particles in a cloud parcel in order to demonstrate the importance of the fact that ice nucleation doesn't stop when cooling ceases. The reasons for this effect are elaborated in [4]. It is difficult to compare our results with other models of ice nucleation in a cloud parcel (e.g., [1], [2], [3]), because those works focus on nucleation by a single substance as opposed to data taken from atmospheric samples. Nonetheless, the underlying physical processes are the same and the different approaches will converge with the availability of more laboratory data and analyses of atmospheric samples.

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