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## 1 INTRODUCTION

This paper describes the basis of an improved ice nucleation scheme for use in a cloud-resolving model (CRM) with bulk-water microphysics. The aim of this scheme is to capture essential characteristics of the evolution of mixed-phase and cirrus clouds, which are due to the limited availability of ice nuclei (IN), whilst retaining the relative computational simplicity of bulk-water microphysics. Results are presented from simulations of an idealised altostratus cloud layer with various warm heterogeneous ice initiation processes active, and a cirrus cloud layer with only homogeneous freezing active.

## 2 HETEROGENEOUS ICE NUCLEATION

Current heterogeneous ice nucleation schemes usually use the Meyer's et al. (1992) combined deposition and condensation-freezing parametrization. This parametrization was derived from IN measurements using a continuous-flow diffusion-chamber, and the number concentration of IN,  $N_d$  (litre<sup>-1</sup>) is given by,

$$N_d = \exp(-0.639 + 12.96\sigma_i) \quad (1)$$

where  $\sigma_i$  is the ice supersaturation (at ice saturation,  $\sigma_i = 0.0$ ). However, in-situ aircraft observations indicate orders of magnitude variability in ice crystal concentrations. When simulating clouds, models sometimes scale  $N_d$  to produce observed ice crystal concentration or ice mass contents.

There are two approaches to using this equation (Khain et al. 2000):

1. The model assumes that the ice crystal concentration at each grid point does not fall below the number of active IN,  $N_d$ .
2. The number of newly activated ice crystals at each time-step,  $dN_d$  is  $12.96N_d d\sigma_i$  if  $d\sigma_i > 0$ .  $N_d$  is still assumed to be the maximum possible value of ice crystal concentration for a given supersaturation.

The first approach can effectively lead to an infinite supply of ice crystals, when ice crystal fall-out removes activated IN from the grid-box, during a subsequent time step the effective IN concentration may be restored by changes to the temperature or ice saturation ratio. In the second approach, problems occur when ice crystals fall from above into the grid box, which has the effect of reducing further ice nucleation.

In such schemes, the maintenance of a stable supercooled mixed-phase cloud is highly dependent on parameter values of the IN scheme. One way to reduce these problems is to utilise an IN aerosol population as a tracer in the CRM, which are converted into ice crystals on activation. These prognostic deposition IN enable ice crystals to be activated once the grid-box goes above a specific ice saturation, and only replenished by entrainment from surrounding unactivated regions.

The UK Met Office Large Eddy Model (LEM) version 2.3 was used for this study. The LEM is a cloud resolving model with three-phase microphysics parametrizations and interactive radiation code. Moist processes are represented by prognostic variables for water vapour, liquid water and ice water mixing ratios. The thermodynamic variable is the potential temperature. The basic state thermodynamic and moisture structure is initialised by the profiles shown in figure 1. The layer in which cloud develops is initially well mixed and ice subsaturated, so that at the start of the

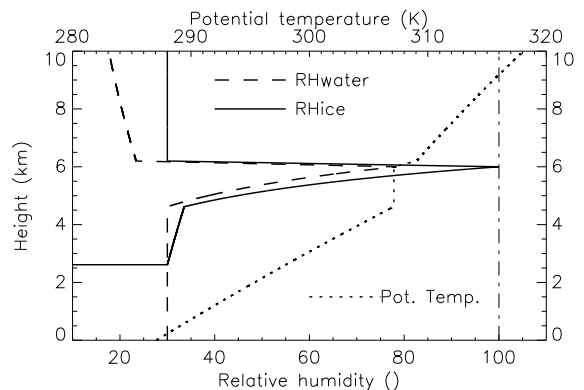


Figure 1: **Initially specified thermodynamic and moisture profiles. The cloud top will be around 6 km and  $-25^\circ$  C**

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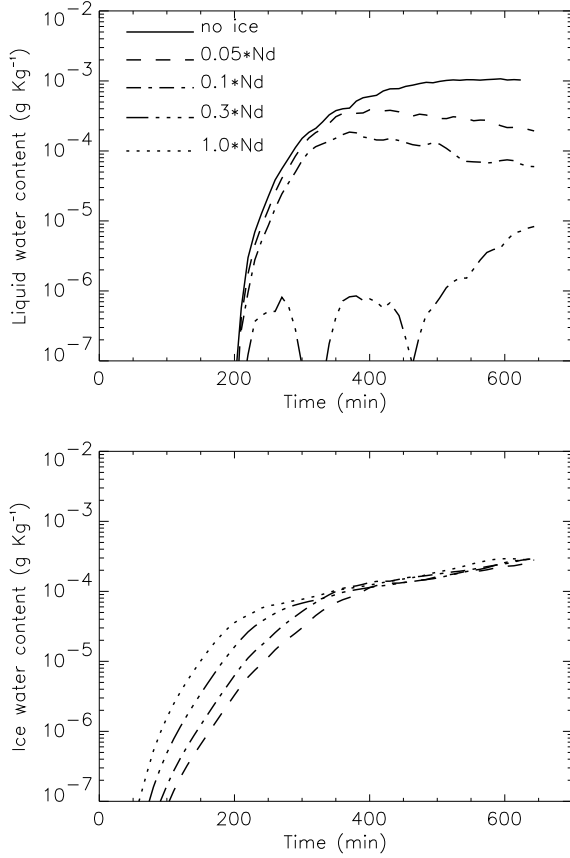


Figure 2: Ice crystals activated up to  $N_d$

model run there are no activated IN, ice crystals or liquid water. Initially the potential temperature is perturbed randomly within  $\pm 0.1\text{K}$ . Cloud top cooling is driven by long-wave radiation effects, solar radiation is switched off.

The domain is cooled to simulate a uniform large-scale vertical motion of  $1\text{--}2\text{ cm s}^{-1}$ . Without domain cooling, mixing of drier air into the cloud layer tends to decrease the cloud water, and makes achieving a steady-state mixed-phase cloud difficult.

The heterogeneous ice nucleation scheme used in the LEM increases the number of activated ice crystals up to  $N_d$  during each time-step according to Equation 1. Results of a mixed-phase altostratus cloud simulation, shown in Figure 2, demonstrate the effect on the liquid water and ice water paths by scaling  $N_d$ . For small changes in the scale factor the cloud evolution changes significantly. Indeed for a scale factor of one, no liquid water is created. A mixed-phase altostratus cloud, which uses a heterogeneous ice nucleation scheme based on increasing the ice concentration up to  $N_d$ , can only be simulated with small  $N_d$  scale factors.

The LEM was modified so that prognostic deposition IN aerosol are used to generate ice crystals rather than the above method. The IN

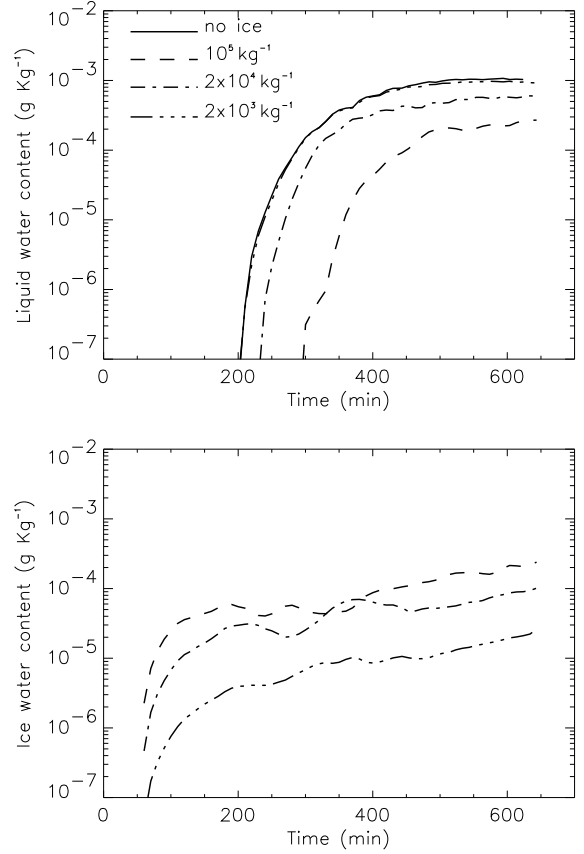


Figure 3: Ice crystals activated using prognostic IN aerosols.

aerosol profile was initially defined so that the concentration mixing ratio was uniform with height. The model runs ranged from  $2 \times 10^3 - 2 \times 10^5\text{ kg}^{-1}$  (around  $1 - 100\text{ litre}^{-1}$  at the cloud level).

For simplicity all IN activate once the grid-box ice supersaturation is above 5%, generating  $1\mu\text{m}$  radius pristine ice crystals. Further ice cannot be initiated until radiatively driven turbulence at the cloud top and bottom entrains unused IN aerosol into the cloud layer. To parametrize an activation spectrum, a series of prognostic IN has to be used, but given the large uncertainty in the actual IN concentrations this approach is needlessly complicated at the present stage. Results are shown in Figure 3.

In the prognostic IN approach, a steady-state mixed-phase cloud can be achieved for a much wider range of IN concentrations. Figure 4. illustrates why this steady-state is possible - the falling ice and snow deplete the liquid water region of IN, enabling the droplets to grow.

Ice crystal activation parametrization is important when the history of the air mass affects the cloud evolution. For example, if models are to simulate stratocumulus where the cloud top is supercooled, but unglaciated, then the IN

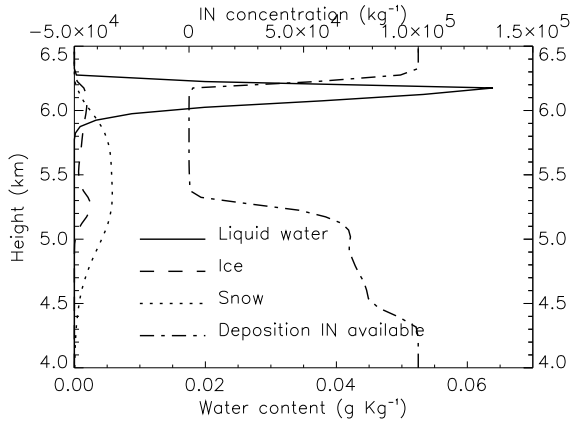


Figure 4: Ice crystals activated using prognostic IN aerosols with  $1 \times 10^4 \text{ kg}^{-1}$  initial number concentration.

must have been depleted upstream by ice crystal fall-out. Any drizzle production will then be from autoconversion and coalescence.

Similarly, prognostic contact aerosol can be used to give a limited supply of contact-freezing activated ice crystals. The aerosol collection rate can include Brownian motion, thermophoretic and diffusiphoretic forcing. The thermophoretic forcing significantly enhances collection during droplet evaporation. It is commonly observed that ice initiation tends to occur in downdrafts of wave clouds (see Field et al. 2001). This can be modelled by prognostic evaporation IN, which are a special subset of CCN that activate when the droplet evaporates.

## 2 HOMOGENEOUS FREEZING OF HAZE PARTICLES

Previous versions of the LEM currently have no ability to generate ice crystals by homogeneous freezing below water saturation, since liquid cloud formation was treated diagnostically. However, it is observed that deliquesced haze particles can freeze homogeneously below water saturation if the temperature is cold enough (at temperatures of  $-40$  to  $-60^\circ$ ).

The homogeneous freezing of supercooled haze particles can be parametrized in the LEM using results from a large number of parcel model runs and prognostic CCN aerosols. The parcel model is an extension of that described in Spice (1999) which considers an adiabatic parcel of air containing a conserved mass of water which rises at a constant updraft velocity. Each parcel is initialised with a different temperature and water relative humidity, these define its total water content.

The homogeneous freezing rate used in the parcel model calculates an effective droplet temperature (which depends on droplet molality), and for highly concentrated haze particles this temperature is high and freezing is inhibited.

Parcel model results show a very rapid onset of homogeneous freezing once a critical relative humidity is reached (see Lin et al. 2002). This critical relative humidity depends on the updraft velocity and parcel temperature. Only some of the haze particles freeze homogeneously before water saturation is reached. The rapidly growing ice crystals then deplete the environmental water vapour and lower the relative humidity. The remaining haze particles are then too concentrated to freeze. Therefore only a fraction of the supercooled haze particles freeze homogeneously. This occurs over a short time interval, which is comparable to the time-step used in CRM's.

This critical freezing relative humidity is plotted as a function of the parcel temperature in Figure 5. Parcel model runs with the same updraft velocity lie on a curve. Varying the updraft velocity generates a family of curves that can be parametrized for use in the LES. Similarly, the final ice crystal concentration from each parcel model run also creates a family of curves, as in Figure 6. Obviously, these curves depend on the CCN chemical species and size spectrum used in the parcel model.

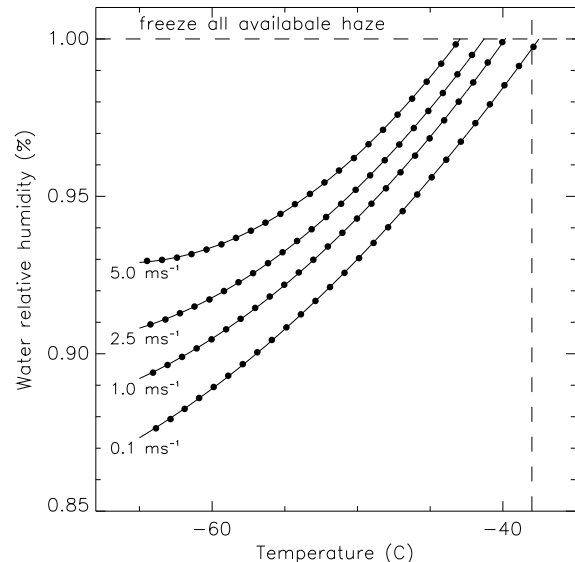


Figure 5: Relative humidity at onset of haze freezing.

The relative humidity thresholds shown in Figure 5. may be parametrized in the LEM, in the form

$$RH_{crit} = a_1 + a_2(T_C + 38.0) + a_3(T_C + 38.0)^2 \quad (2)$$

where the coefficients  $a_1$ ,  $a_2$  and  $a_3$  depend on the updraft velocity, and

$$a_i = b_{1i} + b_{2i}\Gamma + b_{3i}\Gamma^2 + b_{4i}\Gamma^3 + b_{5i}\Gamma^4$$

where the variable  $\Gamma$  is defined by

$$\Gamma = \log \left( -\frac{dT_C}{dt} \right) = \log \left( \text{vert} \frac{g}{C_p} \right)$$

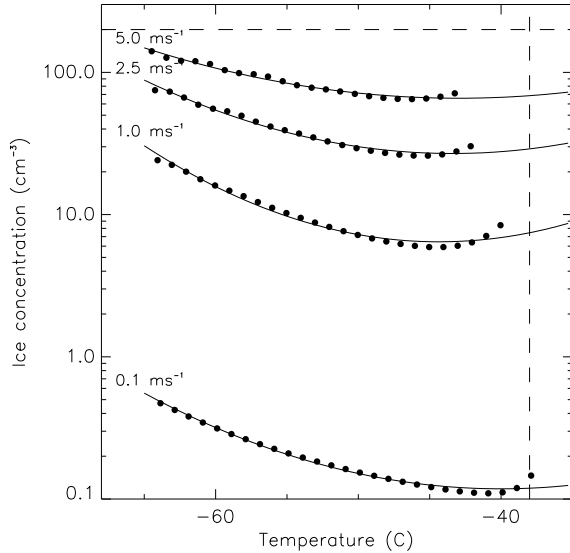


Figure 6: Final ice crystal concentrations.

The activated ice number concentrations shown in Figure 6. is similarly parametrized, in the form

$$\log N_{ice} = c_1 + c_2(T_C + 38.0) + c_3(T_C + 38.0)^2 \quad (3)$$

with the coefficients

$$c_i = d_{1i} + d_{2i}\Gamma + d_{3i}\Gamma^2 + d_{4i}\Gamma^3$$

The LEM then only requires the set of coefficients  $b_{ij}$  and  $d_{ij}$  (one set for each specific CCN chemical species and size spectrum) to determine if freezing has taken place and how many ice crystals are activated.

At each time-step, grid-point values of temperature, humidity and rate of change of temperature (related to the parcel model updraft velocity) are used to determine if the nucleation threshold has been reached. If so, then the ice number concentration is incremented by the value given by Equation 3. and the remaining CCN concentration decremented by the same amount. Results from an initial trial of this scheme are shown in Figure 7. This shows ice created in the updrafts of turbulent cells.

### 3 SUMMARY

The ice crystal activation parametrization has a critical effect on the evolution of mixed-phase clouds. The use of prognostic IN enables a steady-state mixed-phase cloud to develop over a wide range of IN concentrations. A homogeneous nucleation scheme based on a simple prognostic variable for CCN aerosol has also been demonstrated in the LEM. Future case studies using the LEM, will now be capable of using measurements of IN and CCN profiles to control the initiation and evolution of ice and mixed-phase clouds.

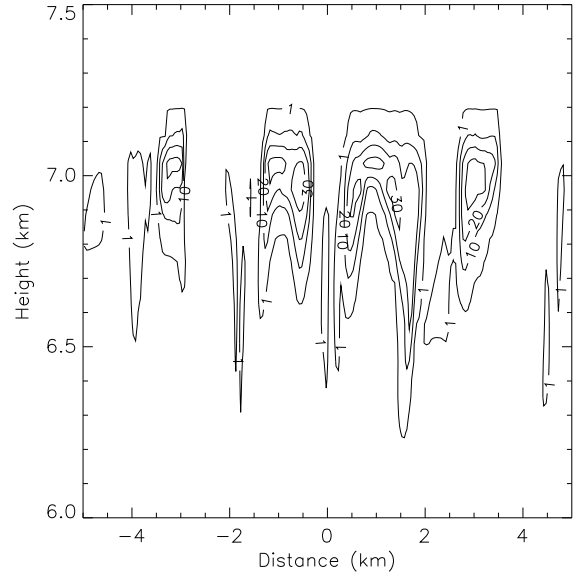


Figure 7: Ice water content ( $\text{mg kg}^{-1}$ ).

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