The development of ice in a cumulus cloud over southwest England

Yahui Huang\textsuperscript{1}, Alan M Blyth\textsuperscript{2,6}, Philip R A Brown\textsuperscript{3}, Tom W Choularton\textsuperscript{4}, Paul Connolly\textsuperscript{4}, Alan M Gadian\textsuperscript{2}, Hazel Jones\textsuperscript{4}, John Latham\textsuperscript{5}, Zhiquang Cui\textsuperscript{1} and Ken Carslaw\textsuperscript{1}

\textsuperscript{1} Institute for Climate and Atmospheric Science, School of Earth and Environmental Science, University of Leeds, Leeds LS2 9JT, UK
\textsuperscript{2} National Centre for Atmospheric Science, ICAS, School of Earth and Environmental Science, University of Leeds, Leeds LS2 9JT, UK
\textsuperscript{3} Met Office, Exeter, Devon EX1 3PB, UK
\textsuperscript{4} School of Earth, Atmospheric and Environmental Sciences, University of Manchester, Oxford Road, Manchester M13 9PL, UK
\textsuperscript{5} National Center for Atmospheric Research, Boulder, CO 80307, USA
E-mail: blyth@env.leeds.ac.uk

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\textbf{Abstract.} An experiment involving the FAAM BAe 146 aircraft, called the ICE and Precipitation Initiation in Cumulus (ICEPIC) project, was conducted in order to measure the microphysical properties of UK summertime cumulus clouds. A line of clouds was penetrated near the ascending tops. Higher concentrations of ice particles than expected from activation on typical ice nuclei using the Meyers formula were observed at relatively high temperatures ($T > -10 ^\circ C$). The observations of numerous ice particles and the coexistence of both small and large cloud droplets, pristine ice columns and graupel pellets within the temperature zone of $-3$ to $-9^\circ C$ strongly suggested the Hallett–Mossop (HM) process of splintering during riming. Agreement between the calculated and observed rates of splinter production supported this suggestion. The Model of Aerosols and Chemistry in Convective Clouds (MAC3) was utilized to establish a quantitative understanding of the observed development of glaciation of this cloud. The results of the model confirmed the important role of the HM process.

\textsuperscript{6} Author to whom any correspondence should be addressed.
They also showed that the warm rain process was fundamental to the production of graupel in the cloud studied, and hence the HM ice particles. A sensitivity test with double the concentration of aerosol particles showed that the concentration of supercooled raindrops decreased as expected, which resulted in fewer graupel particles and a smaller quantity of precipitation, which was delayed by about 5 min. However, the production rate of secondary ice particles generated by the HM process increased due to the increased concentration of small cloud droplets.

1. Introduction

One of the key problems that remains to be solved in cloud physics is to explain quantitatively how precipitation develops in convective clouds through the ice process. Significant precipitation falls from convective clouds throughout the world. Heavy precipitation and flooding events such as occurred at Boscastle, England in the summer of 2004 (Golding et al 2005) are often a result of convection. The most direct reason for the need to understand the physics of the processes that govern the formation and growth of precipitation is to be able to forecast the location, timing and quantity of precipitation. Considerable research has recently been conducted in the US (International H2O Project (IHOP) 2002), Weckwerth et al (2004), the UK (Convective Storm Initiation Project (CSIP), Browning et al (2007)) and Germany (Convective and Orographically-induced Precipitation Study (COPS); Wulfmeyer et al (2008)) to understand the physical processes responsible for the initiation of convection.

Ice particles may begin to form in clouds at temperatures between $-4$ and $-10 \, ^{\circ}\mathrm{C}$ by primary ice nucleation (Cantrell and Heymsfield 2005, Hobbs and Rangno 1985, 1990). They typically initially exist in concentrations of only about $10^3 \, \mathrm{m}^{-3}$ compared with the more numerous supercooled cloud drops whose concentrations can vary from 100 to more than $1000 \times 10^6 \, \mathrm{m}^{-3}$. This balance is critical for the formation of rain as the ice crystals grow and capture the supercooled cloud drops by riming leading to the production of graupel particles. It is also possible that the Bergeron–Findeisen process plays a role in this balance. The basic concepts have been well understood for many years. The challenge is to explain the rates of production and growth of ice and the quantities of ice particles and precipitation particles. Partly, this is because measurements have to be made in the rapidly changing, hostile environment of a convective cloud. The temporal and spatial evolution of the size, shape and concentration...
of the 1 µm to 5 mm particles in concentrations of 10 m$^{-3}$ to 1000 cm$^{-3}$ have to be measured while flying through the cloud at 100 m s$^{-1}$. It is equally difficult for cloud models. It is not yet computationally possible to simultaneously calculate the detailed microphysical processes and the three-dimensional (3D) dynamics found in a typical cumulus cloud with sufficient resolution.

Several studies have shown that in some instances, there is fair agreement between the number of ice crystals that have formed from primary nucleation and the numbers of ice nuclei (Cooper 1986, Mossop 1985a). Meyers et al (1992) found reasonable agreement between observations and their ice nucleation parameterization scheme. There have also been many reports of ice particle concentrations that are much higher than typical concentrations of ice nuclei (e.g. Blyth and Latham 1993, Bower et al 1996, Harris-Hobbs and Cooper 1987, Hobbs and Rangno 1985, 1990, Mossop et al 1972). A number of secondary ice crystal production processes have been suggested to try and explain these observations. Among them, the Hallett–Mossop (HM) process of splintering during riming (Hallett and Mossop 1974) has received the most attention and there is considerable evidence that this process operates in cumulus clouds in many parts of the world. Experiments, reported by Hallett and Mossop (1974) and Mossop and Hallett (1974), showed that ice particles were produced during riming when the temperature was in the range $-3$ to $-8\,^\circ C$. These and subsequent experiments have shown that the process requires both small ($d \leq 13\, \mu m$) and large ($d \geq 24\, \mu m$) cloud drops to coexist with graupel particles (e.g. Mossop 1978, 1985b, Saunders and Hosseini 2001). There is considerable supporting evidence discussed in the above references. The most quantitative analysis has been performed by Harris-Hobbs and Cooper (1987), who found that a predicted rate of splinter production compared well with the observed rate in Florida clouds. Significant time had elapsed between the production of ice particles and when they were measured. The ability to measure smaller particles would allow particles to be imaged and counted closer to where they were produced as in the case discussed by Bower et al (1996) using the holographic imaging system. It should be noted that although there is substantial circumstantial evidence for the HM process operating in clouds, the Harris-Hobbs and Cooper (1987) study is one of the few quantitative results.

An additional factor, first suggested by Chisnell and Latham (1976) and recently supported with results from a Cloud Resolving Model (CRM) and a detailed microphysics model (Phillips et al 2001), is that the rate of production of ice crystals can be substantially enhanced if supercooled raindrops are present. Raindrops that are carried above the freezing level may short circuit the HM process if they freeze in the HM temperature zone. The raindrops may also be a source of secondary ice crystals due to the ejection of ice particles and/or by breaking apart during freezing (Chisnell and Latham 1976, Korolev et al 2004, Rangno and Hobbs 2005). The model results of Phillips et al (2001) in fact showed that raindrops were crucial for the glaciation process. This prediction has not been tested. Columns of supercooled raindrops have been observed in convective cells in Florida by Bringi et al (1997), in New Mexico by Blyth et al (1997) and in England by Caylor and Illingworth (1987).

Additionally, there is difficulty in understanding heterogeneous ice nucleation in clouds (Cantrell and Heymsfield 2005). Significant advances are being made in identifying ice nuclei (e.g. desert dust), in the AIDA chamber (Field et al 2006), but large uncertainties remain, particularly in real clouds. Choularton et al (2008) pointed out the likely importance of the chemical composition of aerosol particles in ice nucleation. They found rapid transitions between highly glaciated regions of cloud and regions consisting almost entirely of supercooled.
water at temperatures of $-11 \, ^\circ C$ for example. They suggested that the transition was caused by ice nucleation initiated by oxidized organic aerosol coated with sulfate in the more polluted regions of the cloud.

In this paper, we present a set of (limited) measurements of the development of ice in a cumulus cloud that formed in a line over southwest England and compare them with the results of a 2D cloud model with sophisticated microphysics (the Model of Aerosols and Chemistry in Convective Clouds (MAC3)). Harris-Hobbs and Cooper (1987) is the only large-scale quantitative study of HM production rates in natural clouds. Hence, similar quantitative studies, such as our present one, are of value in examining whether their conclusions have more general applicability. The aim is to determine the rates of ice production in real clouds and to see how well these can be predicted using the knowledge of the in-cloud thermodynamics and dynamics. In section 2, we describe the details of the observations. Calculations of a predicted ice production rate are compared with an observed rate in section 3. The MAC3 model results are presented in section 4. The summary and conclusions are given in section 5.

All times are given in UTC and altitude as altitude above mean sea level (MSL) unless otherwise stated.

2. The observations

2.1. Details of the experiment

Observations were made on 4 July 2005 in Cornwall, SW England as part of the Ice and Precipitation Initiation in Cumulus (ICEPIC) project. Detailed in situ microphysical measurements were made with the fast forward scattering spectrometer probe (FFSSP), the 2DC (cloud) and 2DP (precipitation) probes (Brenguier et al 1998, Jensen and Granek 2002, Knollenberg 1970, Korolev 2007), and the cloud particle imager (CPI) (Lawson et al 2001) on board the FAAM BAe-146 aircraft. Cloud droplet size distributions for a size range of 1–47 $\mu$m were produced by the FFSSP. Such distributions are highly dependent on the quality of the calibration. Comparisons with hot-wire probes designed to measure liquid water content (Johnson-Williams and Nevzorov probes) suggest that on this flight the FFSSP accurately measured the size range that dominates the liquid water content. The size distribution and concentrations of ice particles were derived from the 2DC and 2DP probes. The nominal size ranges of the 2DC and 2DP, respectively, are 25–800 and 200–6400 $\mu$m. We performed our own analysis of image data from the 2D probes based on standard routines. Since there are known problems with the instrument for sizes smaller than about 100 $\mu$m diameter, only particles with diameters greater than 150 $\mu$m detected by the 2DC and CPI are taken to be ice, based on visual inspection of the images.

Wind measurements were calculated from the inertial navigation system and turbulence probe on the radome in a manner described by Lenschow and Spyers-Duran (1987). Errors in horizontal winds were experienced during turns, as expected, but our interests were only in straight segments of flight through cloud. Vertical wind is thought to have an absolute accuracy within $\pm 0.5 \, m \, s^{-1}$ at best, although the resolution is better than $0.05 \, m \, s^{-1}$. The ports on the turbulence probe occasionally iced up in supercooled cloud, but this did not occur during the penetrations discussed in this paper. An estimation of the altitude of cloud top was made using...
Table 1. Summary of penetrations made during runs 11–14. The variables are as follows: \( t \) is time in UTC; \( z \) is altitude above MSL; \( T \) is temperature measured by the Rosemount probe; \( N_m, L_m, w_m \) and \( N_{i,m} \) are the maximum values during the penetration (run) of, respectively, concentration of cloud drops measured by the FFSSP; liquid water content; vertical wind speed; and concentration of ice particles measured by a combination of the 2DC and 2DP probes, larger than a diameter of 150 \( \mu \)m.

| Run | \( t \) (UTC) | \( z \) (km) | \( T \) (°C) | \( N_m \) (cm\(^{-3}\)) | \( L_m \) (g m\(^{-3}\)) | \( w_m \) (m s\(^{-1}\)) | \( N_{i,m} \) (l\(^{-1}\)) |
|-----|---------------|--------------|--------------|-----------------|-----------------|-----------------|----------------|
| 11  | 113443        | 2.7          | -5.4     | 80              | 0.8             | 6               | 3              |
| 12  | 114120        | 3.0          | -7.0     | 65              | 0.6             | 7.5             | 27             |
| 13  | 114610        | 3.1          | -7.6     | 62              | 0.6             | 5.0             | 42             |
| 14  | 115531        | 3.3          | -8.6     | 120             | 0.7             | 5.0             | 70             |

the onboard forward-looking video camera. Most penetrations were made within a few hundred metres of cloud top.

2.2. Development of ice particles

The aircraft penetrated a system of clouds repeatedly near the tops of new turrets as they gradually ascended. The microphysical parameters measured during these runs are summarized in Table 1. As we will see in more detail below, the clouds encountered a stable layer at an altitude \( z \approx 3 \) km, which will have significantly slowed their ascent, consistent with the observations (Table 1). The eventual top of the cloud system observed is unknown. Some nearby clouds had tops of about 6 km or higher.

The line of clouds were aligned with the wind in an NW–SE direction presumably along a convergence line that formed as a result of a promontory on the NW coast of Cornwall. The sounding launched at 06:00 from Camborne, about 10 km from the clouds studied, is shown in Figure 1. The 0 °C level was at an altitude of about 1.8 km, which is lower than normal for the time of year. The wind direction was from the west to west–north–west and the speed was about 8 m s\(^{-1}\) near the surface, increasing to about 12 m s\(^{-1}\) at an altitude, \( z = 2 \) km. The lifting condensation level (LCL) pressure and temperature calculated from the boundary-layer average values of the potential temperature, \( \theta \), and water-vapour mixing ratio, \( q \), (above the surface layer) are \( p_c \approx 903.63 \) mb and \( T_c \approx 5 \) °C. They approximately agree with the observed values of cloud base pressure and temperature. However, as is usually the case, the water-vapour mixing ratio in the boundary layer is not well mixed. There was a small layer of increased stability near cloud base that the ascending air parcels were able to overcome. The environment was unstable to moist ascent between 900 and 800 mb. There was a stable layer associated with drier air beginning at about 800 mb and then the environment was approximately neutral to moist ascent until about 600 mb. The four penetrations were made near ascending cloud tops between the pressures of 720 and 675 mb.

Figures 2(a)–(d) show information gathered in the four penetrations. During run 11 (Figure 2(a)), the ice particle concentrations were generally less than 2 l\(^{-1}\) and the liquid water content, \( L \), reached 0.8 g m\(^{-3}\). The maximum updraught speed in the centre of the cloud was about 6 m s\(^{-1}\). There were downdraughts on either side of the updraught with speeds of about...
Figure 1. The Camborne temperature and dew-point sounding launched at 06:00.

−1 and −3 m s$^{-1}$ on the left and right sides, respectively. This is frequently found in clouds and is a signature of a thermal (Blyth et al 2005). CPI images (shown on the top of figure 2(a)) were all from cloud drops of diameter from about 30−50 µm. Also a few ice particles were recorded in 2DC and 2DP probes (the images from 2DC are shown in the bottom of figure 2(a)). A few small graupel particles can be seen in both downdraughts. The magnitude of the ice particle concentration in run 11 suggests that there was primary ice nucleation with ice particles in concentrations similar to that estimated with the formula of Meyers et al (1992).

The maximum concentration of ice particles had increased significantly by a factor of about 10 to 27 l$^{-1}$ by the time of run 12 about 5 min later (figure 2(b)). The maximum value of $L$ reduced from 0.8 to 0.6 g m$^{-3}$, whereas the maximum value of $u$ increased slightly to about 8 m s$^{-1}$. There was considerable variability in the ice particle concentration, most likely because of the relatively small sample volume of the 2DC probe. A pristine column crystal was imaged by the CPI on the downshear side of the cloud. Significant concentrations of pristine and rimed columns were also sampled by the 2DC probe in the downdraught on the downshear side of the cloud. A few graupel particles were also observed. There was a larger concentration of ice particles on the downshear side of the cloud.
Figure 2. Ice particle concentration (black line), LWC (blue dashed line) and vertical velocity (red line) for (a) run 11, (b) run 12, (c) run 13 and (d) run 14. Examples of particle images from CPI and 2DC are included. The distance between the vertical lines in the 2DC images corresponds to 800 µm. The size of the particles in the CPI images is shown at the bottom left of the image. Times have been reversed in (b) and (d) so that upshear is on the right in all diagrams and downshear is on the left.

The next penetration (run 13; figure 2(c)) was made about 5 min later at an altitude of about 3.1 km where the temperature was about −7.6 °C. Cloud top had ascended only very slowly, as explained above. The updraught was flanked by downdraughts on either side as in the first
penetration. The maximum concentration of ice particles increased to 42 l⁻¹ in the downdraught at 11:46:05. The maximum value of $L$ was almost the same as in the previous penetration (0.6 g m⁻³), but the maximum updraught speed had decreased to about 5 m s⁻¹. There are peaks in the concentration of ice particles in the two downdraughts. However, there is also a peak in the middle of the updraught. The 2DC images, samples of which are shown in figure 2(c), indicate that the ice particles consisted of small graupel and columns.

The final penetration of this cloud (run 14; figure 2(d)) was made at a temperature of $T \approx -8.6^\circ$C about 9 min after the previous penetration. The maximum concentration of ice particles had increased to about 70 l⁻¹ and the maximum size had also increased. The maximum values of $L$ and $w$ were similar to those in the previous penetration. The 2DC images show that there was a mixture of small and large graupel particles and rimed columns. Irregular ice particles and a column were sampled by the CPI.

In summary, the penetrations were all made near the top of the cloud as it ascended at temperatures ranging from about $-5.5$ to $-9^\circ$C. There are three significant points. Firstly, ice particles were observed in clouds with top temperatures greater than $-6^\circ$C, consistent with the observations presented by e.g. Hobbs and Rangno (1985). Secondly, the concentration of ice crystals in runs 12–14 is greater than the approximately 2 l⁻¹ expected from our current knowledge of ice nuclei (IN) (Meyers et al 1992), although it should be noted that the Meyers et al parameterization is for statistically large ensembles of clouds, and deviations are expected for individual clouds. Also note the recent results of Choularton et al (2008), for example, mentioned in section 1. Finally, the maximum concentration of ice particles increased from about 25 to 70 l⁻¹ in a period of about 10 min. The latter two points suggest that a process of ice multiplication was operating in the cloud.

Figure 3 shows data gathered in run 12 of the concentrations of droplets with diameter, $d > 24 \mu m$ and $d < 13 \mu m$, and the concentration of giant particles with $d > 1$ mm, determined from the images shown in figure 2(b) to be graupel. The figure shows that the maximum concentration of large drops (about 50 cm⁻³) was about an order of magnitude greater than that of small drops. The concentration of small drops was small (the maximum value was 6 cm⁻³), likely because of the clean, maritime nature of the air mass. A significant concentration of graupel was observed. A few graupel particles were also observed in the previous penetration (figure 2(a)). The evidence of abundant ice particles coexisting with pristine columns, graupel particles and large ($d > 24 \mu m$) and small ($d < 13 \mu m$) droplets in the temperature range of $-3$ to $-9^\circ$C suggests that the ice enhancement was due to the HM process of ice splinter production during riming. In the next section, we examine the process quantitatively using calculations of observed and predicted ice production rates outlined by Harris-Hobbs and Cooper (1987).

### 3. Calculations of rate of production of ice

The early laboratory studies of the HM process of secondary ice production due to riming indicated that the ice production rate depended on the number of drops with diameter, $d > 24 \mu m$, accreted by the ice (e.g. Mossop 1976). Subsequent experiments (e.g. Mossop 1978) suggested the importance of accretion of small droplets ($d < 13 \mu m$) as well as larger drops. The hypothesis is that when the larger drops accrete on the graupel surface where small drops have already accreted, there is poor thermal contact with the body of the graupel particle, which allows symmetric freezing of the large drops from the outside inward, resulting in shattering.
Figure 3. Data from run 12, 4 July 2005. The concentration of ice particles (solid line in top panel), vertical velocity (dashed line in top panel), the concentration of droplets larger than 24 µm in diameter (solid line in bottom panel), the concentration of droplets smaller than 13 µm in diameter (dashed line in bottom panel) and the concentration of particles larger than 1 mm in diameter, assumed to be the concentration of graupel (dotted line in bottom panel).

The observed rate of ice production, $P_0$, was calculated according to the formula given by Harris-Hobbs and Cooper (1987):

$$P_0 = \frac{Cd(L_2) - Cd(L_1)}{t_{21}},$$

where $Cd(L)$ is the cumulative size distribution for crystals smaller than $L$. The time for growth between $L_1$ and $L_2$, $t_{21}$, is then given by

$$t_{21} = \frac{(L_2 - L_1)}{G(T)},$$

where $G(T)$ is the average ice particle growth rate from Ryan et al (1976). Harris-Hobbs and Cooper (1987) selected a small size interval of 88–137 µm diameter to minimize the likelihood that ice crystals had grown under conditions differing from those in their current environment and to avoid effects of nearby droplets on crystal growth rates at large crystal sizes. In our calculations, considering no supercooled droplets with diameter greater than 100 µm were found in the data from the CPI, 2DC and 2DP probes for those runs, we selected a size interval of 100–137 µm. We followed Harris-Hobbs and Cooper (1987) and assumed that the ice crystals grew in a water-saturated cloud, that all small ice particles originated from secondary ice production and that new ice crystals were negligible in size, so that by the time they grew to

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Table 2. Comparison of observed and predicted ice production rates.

|         | Run 11 | Run 12 | Run 13 |
|---------|--------|--------|--------|
| Observed rate (l$^{-1}$ s$^{-1}$) | 0.08   | 0.43   | 0.37   |
| Predicted rate (l$^{-1}$ s$^{-1}$) | 0.01   | 0.31   | 0.14   |

Table 3. Values of the function $g$ and coefficient $C$ obtained from three datasets of Mossop.

| Experiment | Function $g$ | Coefficient $C$ |
|------------|--------------|-----------------|
| Q15        | 0.279        | 0.146           |
| Z14        | 0.154        | 0.207           |
| H13        | 0.009        | 0.117           |

A size of about 100 $\mu$m they were similar to those grown in the laboratory experiments of Ryan et al (1976).

A 10 s average was made for each run. Although the average involved part of the downdraft in run 12, the penetration was made near cloud top and the calculation only involved small ice crystals as noted above. The observed ice production rate from the field data gathered in runs 11–12 are shown in table 2 along with the predicted rates calculated below. The observed production rate for run 12 is 0.43 l$^{-1}$ s$^{-1}$, the highest in the three runs. It might have been expected that the highest observed rate of ice production would have been in run 11, because the temperature ($-5.4^\circ C$) was closest to the temperature of the peak splinter rate ($-5^\circ C$). However, the ice production rate in run 11 was only 0.08 l$^{-1}$ s$^{-1}$, which was much lower than the other runs. The pattern of distribution of ice production rate over the three runs reveals that there are two distinct stages of ice production. The low rate in run 11 suggests that the ice development in run 11 is due to primary ice nucleation. The higher rates in runs 12 and 13 (0.37 l$^{-1}$ s$^{-1}$) suggests that the ice development in those runs is due to ice multiplication.

The predicted rate of ice crystal production, $P$, was also calculated from Harris-Hobbs and Cooper (1987), according to the formula:

$$P = C f(T) \int_{R_0}^{R} \int_{r_0}^{r} g(R) \pi (R + r)^2 [V(R) - v(r)] N(R)n(r)E(R, r) \, dR \, dr,$$

(3)

where $R$, $V(R)$ and $N(R)$ ($r, v(r)$ and $n(r)$) are, respectively, the graupel (droplet) equivalent radius, fall speed and size spectrum; $E(R, r)$ is the collection efficiency for graupel–droplet collisions, $C$ is a coefficient that depends on the cloud conditions and $R_0$ and $r_0$ are the respective minimum sizes for $R$ and $r$ that lead to secondary ice production (SIP). The function $f(T)$ is unity at $T = -5^\circ C$ and diminishes linearly to zero at $-3$ and $-8^\circ C$.

The function $g(R)$ is given by

$$g(R) = \int_{r < 6.5} n(r)r^2 E(R, r) \, dr \int_{all} n(r)r^2 E(R, r) \, dr.$$ 

(4)

It represents the approximate fraction of the graupel surface covered by droplets with $d < 13 \mu$m. The value of function $g(R)$ was approximated by the average values obtained from the three experiments of Mossop (1978); these are given in table 3. The value of coefficient $C$ used here was also taken from Mossop (1978) shown in table 3.
The graupel fall speeds of Heymsfield (1978) were used for \( V(R) \) and the graupel density was assumed to be 0.32 g cm\(^{-3}\). The collection efficiency \( E(R, r) \) was taken from Cooper and Lawson (1984). The Stokes fall speeds were used for \( v(r) \). The concentration of graupel was obtained from 2DP data by assuming (based on the images) that particles with diameter larger than 1 mm are graupel.

As seen in table 2, the predicted ice production rate for run 11 is approximately zero. The maximum predicted rate was 0.31 l s\(^{-1}\) found for run 12, while that of the observed rate of ice production from the field data was 0.43 l s\(^{-1}\), also in run 12. The observed and predicted rates for run 13 are 0.37 and 0.14 l s\(^{-1}\), respectively. In addition, the distribution of the predicted rate values over the three runs is similar to that of the observed rate. Taking into account the uncertainties in the calculations that result from the assumptions discussed above, the observed rate of ice production from the field data is in reasonable agreement with the predicted rate of secondary ice production during riming, indicating that the HM process is responsible for the high concentration of ice particles in this cloud.

4. MAC3 model results

In this section, we describe results from the MAC3, a 2D, axisymmetric, non-hydrostatic, bin-resolved cloud model (Reisin et al 1996, Yin et al 2000, 2005, Cui et al 2006). There is a detailed treatment of most major microphysical processes, although the production of ice splinters directly from the freezing of supercooled raindrops (Chisnell and Latham 1976) is not treated. The time and space evolution of size distribution functions is calculated for water drops, ice crystals, graupel and snow particles. The size distribution function for each type of particle is divided into 34 spectral bins with the two-moment approach (number concentration and mass). The model includes cloud drop nucleation and ice particle nucleation, condensation/evaporation (for liquid phase) and deposition/sublimation (for ice phase) and interaction between those particles. There are four modes of ice nucleation in the model: combined scheme of deposition and condensation-freezing (Meyers et al 1992); contact freezing (Meyers et al 1992, Cotton et al 1986); and immersion-freezing (Bigg 1953). The conversion to graupel is important for this study. In the scheme of immersion-freezing in the model, it is assumed that the frozen drops are converted to graupel if the diameter is larger than 200 \( \mu m \). In the parameterization of the process of ice/snow riming into graupel, the conversion is done if the rimed mass is larger than the original mass of ice/snow. Frozen drops are converted to graupel if they collect smaller ice crystals. The model includes detailed calculations of the splinter production during riming (secondary ice production), which was quantitatively described by Hallett and Mossop (1974) and Mossop (1978). The riming rate is directly calculated in the model by solving the stochastic collection equation for drops and ice phase particles with size distributions that are explicitly predicted.

The model domain used for this cloud was 12 km in the vertical and 6 km in the radial direction with a grid size of 300 and 150 m, respectively. The initial environmental conditions used in the model were slightly adapted from the Camborne sounding launched at 06:00 on 4 July 2005 (figure 1). A warm bubble was used to initiate convection in the model.

Measurements of aerosol particles were made with the passive cavity aerosol spectrometer probe (PCASP) in the range of 0.1–3 \( \mu m \). Larger aerosol sizes were obtained from measurements made by the FFSSP and converted to dry sizes. The aerosol size distribution for sizes less than 0.1 \( \mu m \) is taken from the corresponding part of the distribution given by Hobbs.
and Rangno (1985):

\[
dN/d \ln r_n = \sum_{i=1}^{3} \frac{n_i}{(2\pi)^{1/2} \log \sigma_i} \ln 10 \exp \left( \frac{-[\log(r_n/R_i)]^2}{2(\log \sigma_i)^2} \right)
\]

using the parameters provided by Gras (1995). The initial size distribution of aerosol particles used in this study is the combination of the three parts.

Figure 4 shows model results of the spatial distributions of the wind field and the concentration of cloud droplets, ice crystals and graupel at different stages of cloud development. The maximum updraught speed at the core of the cloud was 7.4 m s\(^{-1}\) at \(t = 45\) min and 6.0 m s\(^{-1}\) at \(t = 50\) min. Figures 4(a)–(c) represent the first stage of ice development where primary ice nucleation dominates, figures 4(d) and (e) the second stage where secondary ice production at \(z \approx 3\) km becomes important and figure 4(f) where a combination of the cloud top temperature becoming low enough and the supercooled cloud dissipating leads to the glaciation process being dominated by primary ice nucleation. The aircraft measurements were made through clouds when their tops reached only about 3.5 km, so the third stage was not observed. However, nearby clouds did have higher tops.

At \(t = 35\) min of the simulation (figure 4(a)), ice particles had just been produced at \(z = 3\) km (\(T \approx -8^\circ\)C) and above, up to the top of the cloud and in the downdraught. This is a similar result to that found by Ovtchinnikov et al. (2000) and Phillips et al. (2001) in the simulations of New Mexico convective clouds.

By 40 min (figure 4(b)), the cloud top had ascended to about 5 km, the concentration of ice particles had increased to more than \(51^{-1}\), and the first significant concentration of graupel particles was produced at \(z > 3.5\) km. However, there was no significant graupel in the HM zone (\(-9 \leq T \leq -3^\circ\)C) at \(z \approx 3\) km at this time. Cloud top continued to ascend from 40 to 45 min of the simulation (figures 4(b) and (c)). During this time, the concentration of ice and graupel particles continued to increase. A significant concentration of graupel (0.01 l\(^{-1}\)) had formed at \(z = 3\) km at \(t = 45\) min mainly by freezing of supercooled raindrops. The model results show that the dominant ice production process during this early stage of cloud development is primary ice nucleation.

Cloud top continued to ascend from 45 to 55 min of the simulation and the concentration of ice and graupel particles increased significantly (figures 4(d)–(f)). The cloud reached a maximum altitude at \(t = 55\) min. Notice the increase in the concentration of graupel particles at the lower levels of the cloud as they become larger and the updraught weaker.

As mentioned above, there are four modes of ice nucleation in the model, with deposition and condensation-freezing combined together. Figure 5 shows the ice production rate of the four modes as well as the HM secondary ice production rate at the same times as in figure 4. The figure indicates that deposition/condensation-freezing is responsible for the ice particles near the top of the cloud at \(t = 35\) min (figure 5(a)). As the cloud top ascended to above 4 km, \((T < -15^\circ\)C), immersion-freezing also became significant (figure 5(b)). At 45 min (figure 5(c)), the ice production rate of deposition/condensation-freezing reached more than 100 m\(^{-3}\) s\(^{-1}\) near cloud top, much larger than the rate of immersion-freezing. This means that the newly produced ice particles are mainly very small ice crystals. As a result of the production of graupel particles at 3 km (figure 4(c)), the rate of secondary ice production at \(t = 45\) min reached 17 m\(^{-3}\) s\(^{-1}\). This value was still significantly less than the rate of 26 m\(^{-3}\) s\(^{-1}\) of ice particles produced by primary nucleation.
Figure 4. Spatial distributions of the concentrations of drops, ice crystals and graupel and the wind field at (a) 35 min, (b) 40 min, (c) 45 min, (d) 50 min, (e) 55 min, and (f) 60 min of cloud simulation. The blue lines are the concentration of liquid drops with contours at 1, 30, 50 and 100 cm$^{-3}$; the red lines are the concentration of ice crystals with contours at 1, 5, 20, 30 and 50 l$^{-1}$; and the green lines are the concentration of graupel particles with contours at 0.01, 0.1, 0.5 and 1 l$^{-1}$. The scale of the wind vectors is shown at the top of (a).
Figure 5. Ice nucleation rates of immersion-freezing, deposition/condensation-freezing and contact-freezing and secondary ice production rate of the HM process. The contours are 0.1, 1, 50, 100 m$^{-3}$ s$^{-1}$, etc. The dashed lines in the figure represent isotherms in °C.
There was a marked increase in the concentration of ice particles at $z = 3$ km after 45 min—see figures 4(d)–(f) and 5(d)–(f). The maximum concentration of large and small droplets at $z = 3$ km and $t = 45$ min was 37 and 23 cm$^{-3}$, respectively, and was 20 and 10 cm$^{-3}$, respectively, at $t = 50$ min (not shown). The HM process of splintering during riming was active in the model at those times. As a result, the concentration of ice crystals increased at $z = 3$ km from 2.5 to 38.5 l$^{-1}$ in just 5 min. Figure 5(d) shows increased values of the secondary ice crystal production rate at $t = 50$ min. The maximum value was 0.47 l$^{-1}$ s$^{-1}$ at $t = 50$ min at $z = 3$ km. The maximum concentration of ice particles at $t = 55$ min, $z = 3$ km continued to increase to 63.8 l$^{-1}$ (figure 4(e)). However, the ice crystal splinter production rate decreased to 0.36 l$^{-1}$ s$^{-1}$ (figure 5(e)) which resulted in a decrease in the concentration of ice particles at $z = 3$ km at $t = 60$ min (figure 4(f)). Figure 5(f) shows that the ice crystal splinter production rate decreased further at $t = 60$ min. This was due to the lack of cloud drops.

The time evolution of the microphysical parameters can be seen in more detail in figure 6. The diagram plots the maximum model values of parameters at a given altitude and time. The concentrations of cloud drops, $N_d$, increased to a peak value of 130 cm$^{-3}$ (figure 6(a)). The values of liquid water content, $L$, and vertical wind speed, $w$, (not shown) all increase uniformly together with cloud top to peak values of 1.8 gm$^{-3}$ and 8.5 m s$^{-1}$, respectively, at $t = 36$ min. The values of $N_d$ and $w$ decreased thereafter. The values of $L$ fluctuated presumably due to entrainment events before decaying in a similar manner to the values of $N_d$ and $w$.

The first values of the concentration of ice crystals at a given altitude, $N_i$, increased gradually as cloud top ascended. Notice that there is a peak of about 50 l$^{-1}$ commencing at about 50 min at $z \approx 3$ km. This is due to the secondary production of ice splinters by the HM process. The peak value of $N_i$ reached about 142 l$^{-1}$ at $z \approx 6$ km ($T \approx -29^\circ C$) at $t = 63$ min.

Figure 6(c) shows that the value of the maximum concentration of graupel particles, $N_g$, first reached 0.01 l$^{-1}$ at $t = 40$ min and $z \approx 4$ km near cloud top. The value of $N_g$ increased to 1.7 l$^{-1}$ at $z \approx 5$ km and $t = 49$ min. However, we show below that it is the graupel particles that were formed by the freezing of supercooled raindrops at $z \approx 3$ km that are crucial for the secondary production of ice by the HM process. The model run can be compared with the observations despite the model cloud top being 6 km instead of 4 km, because the particles created at upper levels have little influence on the production of HM ice particles at lower levels.

Graupel particles can be generated in the model by the growth of ice crystals through deposition followed by riming, or by short-circuiting that process by directly freezing supercooled raindrops. One mechanism for this freezing is by collisions between small ice crystals and the supercooled raindrops (Phillips et al 2001). A small concentration of supercooled raindrops (defined as drops with $d > 1$ mm) formed by collision and coalescence before the graupel particles at $t = 37$ min and $z = 2.7$ km, about 1.5 km above cloud base. This altitude is similar to that found by Lowenstein (2007) for shallow, maritime trade-wind cumulus clouds, with similar concentrations of cloud droplets. The supercooled raindrops ascended to a maximum altitude of about 5 km. Figures 7 and 8 show the spatial distributions of raindrop mass and graupel particle mass, and the size distributions of drops and graupel, respectively, both for 40 and 45 min. It is evident from the figures that graupel particles formed at about the same time and altitude as the supercooled raindrops (figure 7(a)) and the mass of graupel particles increased at the same time as the increase in mass of supercooled raindrops (figure 7(b)). Furthermore, the size distributions of the graupel particles at 40 and 45 min (figure 8) show that the graupel has formed from freezing the supercooled raindrops, because of the large size.
Figure 6. The evolution of the model-produced values of (a) the concentration of cloud droplets, (b) the concentration of ice crystals, (c) the concentration of graupel particles, (d) the reflectivity and (e) the concentration of raindrops. Each diagram shows the maximum model values at a particular altitude and time.
Figure 7. The spatial distributions of raindrop mass and graupel mass at (a) 40 min and (b) 45 min. The solid lines represent the raindrop mass and dashed lines the graupel particle mass.

Critically, the graupel that exists at \( z \approx 3 \) km formed as a result of raindrop freezing. The drops freeze as a result of collision with small ice crystals or by immersion-freezing. Therefore, the graupel particles were not formed by the slower process involving the growth of ice crystals followed by riming, because they exist soon after the appearance of the first ice, they form at the same altitude and time as the raindrops and their initial size is large. In any case, all the graupel particles at \( z = 4.2 \) km at \( t = 45 \) min had terminal velocities less than the updraught speed (7 m s\(^{-1}\)), except for a very small number of large particles.

Figure 8. The size distributions at 40 min (left) and 45 min (right). The solid lines represent liquid drops and the dashed lines graupel particles at 3.6 km in the cloud core.
Table 4. Summary of comparison of model results with observations. The maximum values of LWC and concentration of ice crystals are given. $w_{mx}$ and $w_{mn}$ are the maximum and minimum values of the vertical velocity, respectively. The following key is used for model altitudes and times. Level 1: $z = 2.7$ km; $t = 45$ min; level 2: $z = 3$ km; $t = 50$ min; level 3: $z = 3.3$ km; $t = 55$ min. Note that the top eventually reached by the cloud observed is unknown.

| Parameter | Observations | MAC3 results |
|-----------|--------------|--------------|
| Geometry aspects | | |
| Cloud base alt (km) | About 1.0 | About 1.0 |
| Cloud top alt (km) | – | 6.0 |
| Cloud width (km) | 3.0–4.0 | 3.0–4.0 |
| Vertical velocity (m s$^{-1}$) | | |
| Run 11: | Level 1: |
| $w_{mx} = 6.0$, $w_{mn} = -3.0$ | $w_{mx} = 6.0$, $w_{mn} = -0.5$ |
| Run 12: | Level 2: |
| $w_{mx} = 7.0$, $w_{mn} = -2.0$ | $w_{mx} = 5.3$, $w_{mn} = -0.6$ |
| Run 14: | Level 3: |
| $w_{mx} = 5.0$, $w_{mn} = -4.0$ | $w_{mx} = 4.1$, $w_{mn} = -0.2$ |
| LWC (g m$^{-3}$) | | |
| Run 11: | Level 1: |
| 0.8 | 1.2 |
| Run 12: | Level 2: |
| 0.6 | 0.7 |
| Run 14: | Level 3: |
| 0.7 | 0.3 |
| Conc of ice crystals (l$^{-1}$) | | |
| Run 11: | Level 1: |
| 3.0 | 1.4 |
| Run 12: | Level 2: |
| 27.0 | 38.5 |
| Run 14: | Level 3: |
| 70.0 | 55.1 |
| Ice production rates (l$^{-1}$ s$^{-1}$) | | |
| Run 11: | Level 1: |
| 0.08 | 0.01 |
| Run 12: | Level 2: |
| 0.43 | 0.47 |
| Run 13: | Level 3 (approx.): |
| 0.37 | 0.36 |

It is clear therefore that the second stage of ice development is caused by HM ice particles when the graupel is formed from freezing the supercooled raindrops within the HM temperature zone. The values of $N_g$ lie between 0.1 and 0.5 l$^{-1}$ at $z \approx 3$ km between 50 and 60 min when there is a peak in the values of $N_i$.

The first 10 dBZ Rayleigh reflectivity echo appeared at $z \approx 3.5$ km at $t \approx 36$ min (figure 6(d)), produced by the supercooled raindrops. The reflectivity increased thereafter to a maximum value of 61 dBZ at $z \approx 2$ km, below the peak in concentration of graupel. There is evidence, in the lowest 500 m after $t \approx 52$ min of figure 6(e), of the graupel particles melting before they hit the ground.

4.1. Comparison with observations

Three model levels and times of $(z, t) = (2.7$ km, 45 min), (3 km, 50 min) and (3.3 km, 55 min) were selected for comparison with the observations corresponding to the altitudes of runs 11, 12 and 14, respectively—see table 4. Run 13 and level 3 were used for comparison of the ice production rates. There is reasonable agreement between most model variables and observations with a few exceptions. Although the model predicted that supercooled raindrops were formed...
by the warm rain process and that they played an important part in the production of ice, no such drops were observed in the cloud. The model predicted that the concentration of supercooled raindrops was no greater than 0.31 l−1, so it is possible that they were present but not observed by the probes which have low sample volumes. The model values of cloud liquid water content are higher than the observed values and the downdraught speeds are too low. This is a common problem in cloud models. In order to have a semi-realistic treatment of turbulent entrainment of non-cloudy air into the cloud, the model should be 3D with a resolution of about 10 m or less. It would then not be possible to include such detailed microphysics.

The most important variables for this study are the concentration of ice particles and the HM production rate. Graupel is more difficult to measure because of the relatively small concentrations and the small sample volume of the probes. But the HM production rates depend on the concentration of graupel. It can be seen from table 4 that the model values of ice particle concentration are remarkably similar to the observed values for the three penetrations. Not surprisingly, the model HM ice production rate agreed well at (3 km, 50 min). This value also agrees with the predicted rate of 0.31 l−1 s−1 calculated from equation (3).

4.2. Sensitivity to doubling of aerosols

In this section, we describe the results of a sensitivity model run where the concentration of aerosol particles was doubled. The control and double-aerosol runs are henceforth called A1 and A2, respectively. The supercooled raindrops produced in the model had a significant influence on the formation of graupel and hence on the concentration of ice particles as a consequence of the production of ice splinters during riming (the HM process). The formation and evolution of raindrops depend on the aerosol size distribution (Cui et al 2006, Lowenstein 2007). An increased concentration of aerosol particles can result in a reduced number of raindrops produced by collision and coalescence due to the decreased size of the larger number of cloud drops. Figure 9 shows the evolution of the concentrations of drops with diameter, \( d < 10 \mu m \), drops with \( 50 < d < 63 \mu m \), raindrops (assumed here to be cloud drops larger than 1 mm), and graupel particles for runs A1 and A2. The values shown in the figure were the totals over the whole depth of the cloud. More small cloud drops were activated in run A2 (figure 9(a)) and fewer large drops developed (figure 9(b)) because of competition for the available water vapour (Beard and Ochs 1993, Squires 1958). Since growth by collision and coalescence depends on the size of the drops, the smaller concentration of 50 \( \mu m \) and larger drops suggests that a smaller number of raindrops will be produced. Figure 9(c) shows that there is indeed a reduction in the total concentration of raindrops in run A2, which agrees with previous work (Cui et al 2006). As a consequence, the concentration of graupel is also reduced (figure 9(d)) since some graupel is formed by the freezing of supercooled raindrops.

According to Hallett and Mossop (1974) and Mossop (1978), the rate of ice multiplication of the HM process depends not only on the concentration of graupel particles available, but also on the presence of large (\( d > 24 \mu m \)) and small (\( d < 13 \mu m \)) cloud droplets. The concentration of small drops was increased in run A2 (figure 9(a)). Despite the decrease in concentration of supercooled raindrops and hence graupel particles in the HM temperature zone, the number of ice particles produced in the HM zone in run A2 was approximately doubled (figure 10(b)). The rate of splinter production in run A2 at 55 min was 0.98 l−1 s−1, more than double the maximum rate of 0.47 l−1 s−1 in run A1. The concentration of ice crystals also increased up to an altitude of about 5 km from about 53 to 65 min.

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Figure 9. Evolution of the concentrations of (a) drops with diameter less than 10 \( \mu \text{m} \), (b) drops with \( 50 < d < 63 \mu \text{m} \), (c) raindrops and (d) graupel particles. In the figure, the solid lines represent the results from the control run and the dashed lines the results from the experiment with double aerosol loading.

As expected, the other principal difference between runs A1 and A2 is the timing of the production of raindrops and graupel particles. As can be seen by comparing figures 6 and 10, the first particles were produced about 4 min later in run A2. The maximum instantaneous rain rate decreased from 92.7 to 83.8 mm h\(^{-1}\). There were no other substantive changes to the behaviour of the cloud.

5. Summary and conclusions

In this paper, we have described the in situ microphysical measurements made by the FAAM BAe 146 aircraft on 4 July 2005 during the ICEPIC field campaign and the MAC3 model results. The aircraft penetrated a single cloud as the cloud top ascended from a temperature of \(-5^\circ\text{C}\) to \(-9^\circ\text{C}\). During this period, the observations and modelling showed that there was a two-stage ice-forming process. Initially, low concentrations of ice particles were found with values comparable with that estimated with the formula of Meyers et al (1992) derived from typical IN measurements. In the second stage, the concentrations of ice particles increased to values greater than could be explained by typical concentrations of IN. The observations of numerous ice particles and the coexistence of small \((d < 13 \mu \text{m})\) and large \((d > 24 \mu \text{m})\) cloud droplets, pristine ice columnar habits and graupel pellets within the temperature zone...
Figure 10. Same as figure 6, but for increased concentration of droplets.

of $-3$ to $-9^\circ$C strongly suggested that the HM process of splintering during riming was responsible for this increase in the concentration of ice. This was confirmed by comparing the predicted rate with the observed rate of ice particle production (Harris-Hobbs and Cooper 1987, Rangno and Hobbs 1994).

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We also simulated the formation and growth of ice particles using MAC3—a 2D axisymmetric cloud model with bin-resolved microphysical processes. The model results were in reasonable agreement with observations, which further confirms that the HM process can account for the generation of the observed high concentration of ice particles. The agreement does not rule out the possibility that some of the observed ice particles were produced directly as a result of the freezing of supercooled raindrops (Chisnell and Latham 1976), but clearly they could not have dominated the splinter production process at the stage when this cloud was observed.

The MAC3 simulations strongly suggest how the second stage of ice development likely occurred in the observed clouds. Supercooled raindrops are formed via the warm rain process. They freeze and become graupel particles that can collect the large and small cloud drops. Splinters are produced in the HM temperature zone. The formation of supercooled raindrops is key in this cloud, because they short circuit the process of production of graupel. The cloud has dissipated by the time the graupel, formed through the growth by diffusion and then riming of ice crystals, reach the HM zone. No supercooled drops were observed, however. It is possible that the concentrations were below the detectable limit of the instruments.

A sensitivity test was performed with double the concentration of aerosol particles ingested at cloud base to investigate the effect on the glaciation process and development of precipitation. The results showed that for this cloud, the amount of warm rain decreased and was delayed as expected, which resulted in fewer graupel particles and a smaller quantity of precipitation, which was delayed by about 5 min. Despite the reduction and delay in production of supercooled raindrops and decrease in concentration of graupel particles, the production rate of secondary ice particles generated by the HM process increased due to the increased concentration of small cloud droplets.

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