

Stochastic ice nucleation in supercooled clouds, and constraints on the fraction of small ice crystals in glaciated clouds, observed using Doppler lidar and radar

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ABSTRACT

Using vertically-pointing Doppler lidar and Doppler radar measurements, we show that 1) the number of crystals smaller than 100 μm in ice-phase clouds is much lower than measured in-situ by cloud droplet probes, likely due to crystal shattering on the probe inlet; and 2) that the nucleation of ice in persistent supercooled layer clouds is a stochastic process, leading to a steady flux of ice particles over the course of many hours.

1. SMALL CRYSTALS IN ICE CLOUDS

There has been a great deal of controversy over the concentration of ice crystals smaller than $\approx 100\mu\text{m}$ in size which are present in natural clouds; conventional in-situ 2D shadow probes do not sample these small particles accurately, and forward scattering cloud droplet probes such as FSSP [1] are often used as a substitute for sampling these small particles [2]. Such studies have reported extremely large concentrations of sub-100 μm ice crystals, up to 10^4 /litre [3], whilst concentrations of larger crystals measured by 2D shadow probes are typically only 1-10/litre [4]. If genuine, the enormous numbers of small crystals observed by the FSSP would exert a substantial influence on the radiative properties of ice-phase clouds.

On the other hand, there is evidence that FSSP concentrations in ice clouds may be strongly affected by artefacts [5-9], caused by crystals and snowflakes shattering on the probe inlet, leading to numerous satellite ice fragments, which are subsequently counted by the FSSP detector.

Typically [10,11] general circulation models (GCMs) represent ice particles using a simple gamma distribution, with concentration N of particles with diameter D given by:

$$N = N_0 D^\mu \exp(-\lambda D) \quad (1)$$

where one of the parameters λ or N_0 is diagnosed as a function of temperature, and the other is predicted from the model ice water content (IWC). The parameter μ is usually fixed to be a constant value ($=0$ for a simple exponential). However, if the measured concentrations of small crystals are genuine, equation (1) substantially underrepresents their numbers.

To represent the small particles, Ivanova *et al.* [12] have analysed FSSP and 2D shadow probe data from 17 flights through mid-latitude stratiform ice clouds and use them to parameterise a two mode ice particle size spectrum for use in numerical weather and climate

models. They found that the size spectra could be well described by adding a second narrow gamma distribution, centred around $D=25\mu\text{m}$, onto equation 1: examples of this parameterisation at various temperatures are illustrated in figure 1. In this distribution particles 25 μm in size are 2-3 orders of magnitude more numerous than particles 250 μm in size. Mitchell *et al.* [13] applied this bimodal parameterisation to a GCM and found that the large numbers of tiny crystals had a significant impact on ice cloud coverage and radiative forcing. It is therefore vital to determine whether these large numbers of crystals are genuine or not, if simulation of ice cloud in numerical weather and climate models, and their associated radiative impacts are to be simulated realistically.

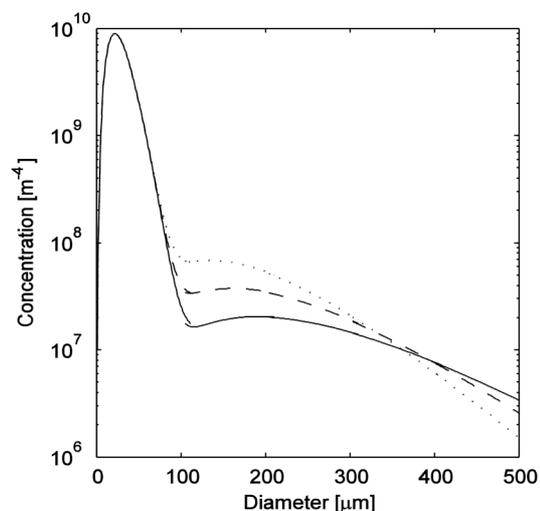


Figure 1: Example ice particle size spectrum for $T=-15$ (solid), -25 (dashed) and -35C (dotted). The IWC is fixed at $0.01\text{g}/\text{m}^3$.

1.1 Doppler Lidar measurements

Measurements were made using a vertically pointing 1.5 μm Doppler lidar at the Chilbolton observatory in Hampshire, UK. The instrument has a maximum range of 10km, and measures profiles of backscatter and Doppler velocity every 32s at 36m resolution. The observations presented here were collected continuously between September 2006 and January 2008. Liquid clouds, boundary-layer aerosol and ice cloud contaminated by specular reflection from oriented plate crystals are removed using the methods described in [14]. This processing effectively limits the dataset to strati-

form ice clouds which are not precipitating at the ground. 1 million 32sx36m ice-phase pixels were analysed.

For lidar measurements at non-absorbing wavelengths an ice particle with a given shape and orientation produces a backscatter proportional to its projected area $A(D)$, and the corresponding Doppler velocity would represent the area-weighted fall speed of the crystal population. However, at $1.5\mu\text{m}$ there is some absorption as the light reflects around the inside of the crystal: as the particles become larger, this absorption increases, reducing the backscatter. This leads to the Doppler velocity being more strongly weighted towards the smaller particles than a simple area-weighting:

$$\langle v \rangle = \frac{\int N(D)A(D)f(D)v(D)dD}{\int N(D)A(D)f(D)dD} + w \quad (2)$$

where v is the particle fall speed, w is the vertical wind, and the factor $f(D)$ falls off as a function of crystal size from $f \approx 1$ for small crystals, to $f \approx 0$ for very large ones.

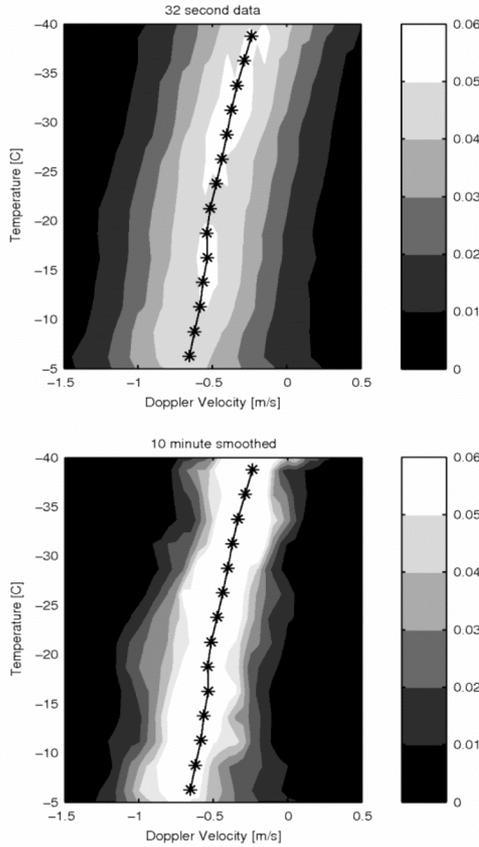


Figure 2: Distribution of lidar Doppler velocity in ice cloud as a function of temperature, at 32s resolution (top) and smoothed over 10mins (bottom). Distributions are accumulated from 17 months of continuous measurements.

Figure 2 shows the distribution of Doppler velocity for all the ice cloud sampled by the lidar over the 17 month period as a function of temperature. The distribution is broad, and has a clear trend with particles falling faster at warmer temperatures, indicating the influence of particle growth/aggregation. Note the sign

convention of negative Doppler velocities for particles falling toward the lidar. Smoothing the data over 10 minute periods before calculating the statistics leads to a narrower distribution also shown in figure 2, as small scale variations in w are removed. We expect any large scale ascent to be weak relative to the ice crystal terminal velocities.

We now investigate whether our observed Doppler measurements are compatible with Ivanova *et al*'s bimodal size spectra.

1.2 Comparison with forward model

We took Ivanova *et al*'s parameterisation (figure 1) and calculated the associated terminal velocities of the particles v using the method of Mitchell [16]. We note that λ (and therefore $\langle v \rangle$) is diagnosed from the cloud temperature, so there is no dependence on IWC to consider. The size spectra were integrated over the size distribution with an area weighting to compute $\langle v \rangle$ as per equation (1) with $f=1$ and $w=0$. Various mass-size and area-size relationships were used for the ice particles to test the sensitivity of the forward model. The results are shown in figure 3a, alongside the mean *observed* Doppler velocity, binned by temperature. Whereas equation (3) leads us to expect that the forward modelled velocities using $f=1$ should be too fast relative to the $1.5\mu\text{m}$ observations, the comparison in figure 3a shows the opposite – the velocities from the bimodal size spectrum are much slower than observed, by a factor of two at warm temperatures. This is evidence that the small crystals (which fall at only a few cm/s) are exerting a much stronger influence on the parameterised spectra than they do in the clouds we observed.

One concern about the above conclusion is that it relies on the balance between the small and large crystal modes. It may perhaps be that Ivanova *et al.* included too few large crystals, instead of too many small ones. To test this, we have calculated the mean Doppler velocity observed by coincident 35GHz Doppler radar measurements in the same ice clouds, figure 3d. The radar is sensitive only to the larger particles because of the mass² weighting of Rayleigh scattering, and therefore provides a test of the large mode. This shows that if anything, there are *too many* large particles in Ivanova *et al*'s large mode, rather than too few. This re-enforces our conclusion about the small crystals.

Removing the small crystal mode from the parameterised spectra brings the observed and forward modelled curves into much closer agreement as shown in figure 3c. Reducing the small mode by a factor of 5 also gives reasonable agreement with observations (figure 3b) – in such a case the contribution from the small crystals is only 10% of the cloud's total projected area.

We conclude that the large numbers of small crystals observed by FSSP in-situ are not compatible with our new Doppler lidar measurements of a large sample of ice clouds, and that they should not be included in numerical weather and climate models. This work is described in more detail in reference [15].

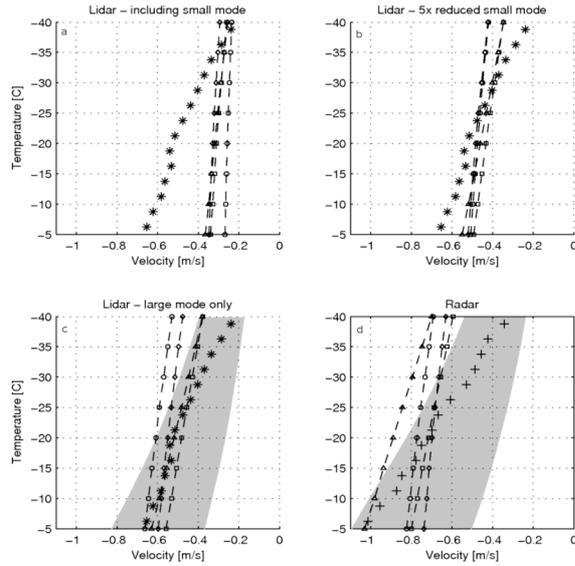


Figure 3: Comparison between forward modelled (lines with symbols) and observed mean Doppler velocities as a function of temperature. Symbols indicate mass-area-diameter relationships: square=planar polycrystals, triangle=rosettes, diamond=aggregates, circle='cold type'. Grey shading shows forward model of Wilson and Ballard (1999) scheme for reference ($IWC=0.001-0.1g/m^3$).

2. ICE FORMATION IN SUPERCOOLED LAYER CLOUDS

The freezing of liquid water droplets to form ice in supercooled clouds is a key process for the formation of precipitation, and the subsequent depletion of liquid water via the Bergeron-Findeison mechanism has an important influence on the cloud's interaction with radiation. One fundamental question of great importance to how ice nucleation is represented in numerical models is whether it is a *singular* process where each nucleus (if present in the droplet) freezes immediately once cooled to a specific temperature, or a *stochastic* process like radioactive decay, where there is a random element to the freezing, ie where many droplets contain a potential nucleus, but each nucleus individually has a small probability of freezing in a given time interval at a given temperature.

Lidar and radar are valuable instruments for probing supercooled clouds: the lidar is very sensitive to the presence of liquid water droplets (because they are so numerous) whilst the radar return is dominated by the ice crystals which are much larger. An example set of observations is shown in figure 4, sampled on 18 May 2008. A layer of supercooled liquid water is highly visible as a thin strip of strong backscatter at the top of the cloud (4km), with much weaker backscatter in the ice-phase virga beneath. There are also some transient liquid layers embedded in the virga. This lidar points a few degrees off vertical; a second instrument pointing directly at zenith shows a radically altered picture, with similar backscatter in the liquid layer, but much stronger (1 order of magnitude) backscatter in the virga. This is the result of specular reflection from plate-like crystals which have been nucleated and

grown in the liquid layer. The cloud top temperature is -15C.

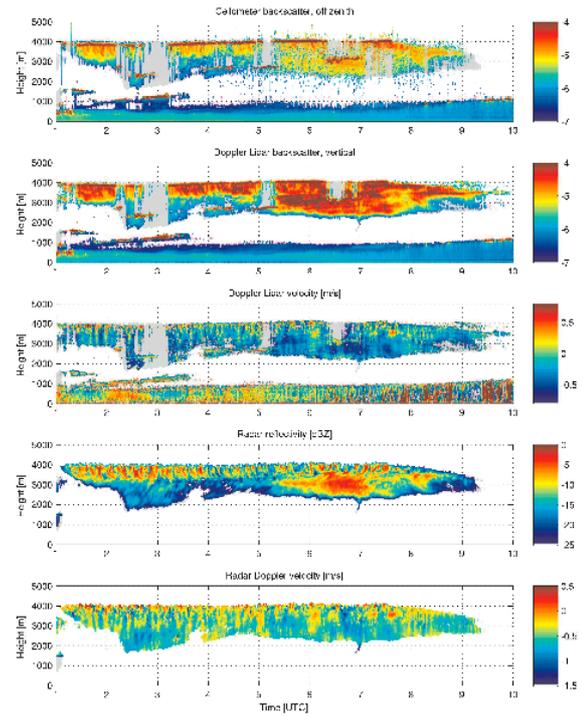


Figure 4: (top down) backscatter from lidar pointing 4° off zenith; backscatter from lidar pointing directly vertical; lidar Doppler velocity (vertical); coincident radar reflectivity; radar Doppler velocity. 18 May 2008.

The cloud radar time series shows the reflectivity from the ice in the top 500m of cloud appears to be fairly steady throughout the observing period, modulated on small scales by size-sorting into fallstreaks; lower down in the virga the reflectivity is much variable, due to the changing humidity profile. The Doppler measurements from lidar and radar show small scale convective overturning in the liquid layer as the cloud is cooled radiatively from the top; also present are longer period waves. These vertical motions likely help maintain the liquid cloud.

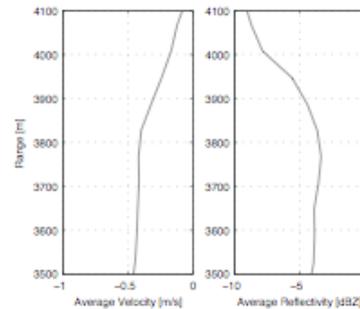


Figure 5: mean Z, V profiles at top of cloud 18 May 08.

Figure 5 shows the profile of radar reflectivity and Doppler velocity in the top 600m of cloud, averaged over the complete 10hr period. Most of the growth occurs in the top 300m (the supercooled layer), below which the profiles are flat. The reflectivities suggest an ice water content of a few hundredths of a gram per m^3 , and therefore a significant flux of ice crystals, which is maintained for many hours. The cloud top is

quite flat over this time period and radiosonde ascents suggest the temperature varies by less than 1 degree. The implication is that a) the ice crystals are being nucleated at a steady rate through the observing period, ie a stochastic process, or b) fresh singular ice nuclei are being mixed into the cloud at a steady rate. Based on the thickness of the layer we estimate LWP $\sim 40\text{g/m}^2$; the air above the layer is extremely dry.

A quantitative estimate of the flux of ice from these clouds is provided by a second case study from Chilbolton, on 18 Feb 2009. In this second case study the FAAM 146 aircraft was sampling in the vicinity of Chilbolton as part of NERC's *APPRAISE-CLOUDS* project. The radar (figure 6) recorded a similar situation to that on the 18 May 08 case, with a long-lived mid-level layer cloud precipitating ice. However low-level stratocumulus was also present, obscuring the lidar beam. Radiosonde ascents and in-situ sampling confirmed the presence of a thin layer of liquid water at the cloud top, with an LWP $\sim 40\text{g/m}^2$. Cloud top was -13C .

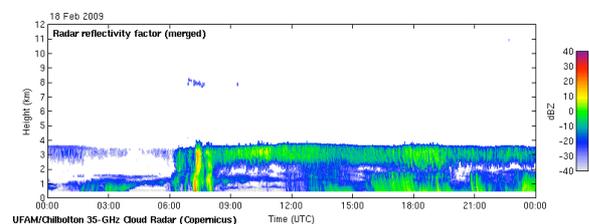


Figure 6: *Cloud radar observations 18 Feb 2009.*

Again, the radar shows an approximately constant flux of ice from the supercooled cloud, maintained for many hours. Again we observed dry air overlying the cloud, with a temperature inversion separating the two air masses. The same arguments about how this ice is nucleated therefore apply to both case studies. The coincident aircraft sampling allows us to be more quantitative in this second case: the size spectrum from measured by 2D shadow probes on the aircraft were integrated with a number of different velocity-diameter relations (different symbols) to estimate the flux of ice falling out the supercooled layer: see figure 7. The flux is $\sim 100\text{m}^2/\text{s}$ or 1 million crystals per m^2 every 3 hours. This continues for ~ 20 hrs.

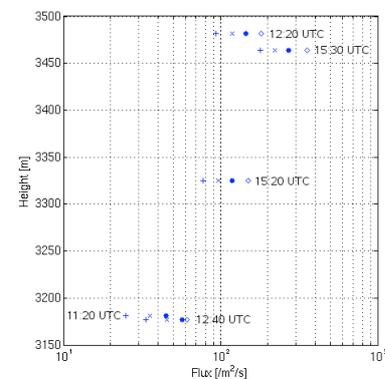


Figure 7: *Flux of ice falling from supercooled layer 18 Feb 2009, estimated from 2D in-situ probes.*

Typically [17] the number of singular nuclei at -13C is $1/l$. In the 300m thick liquid layer these nuclei would be

completely exhausted in 3hrs. If the process is stochastic the implication is that only 1 in 10^5 droplets freeze per hour (if all droplets contain a potential IN). Stochastic freezing therefore appears to be key to ice production in supercooled layers, but is very difficult to measure in the lab because of the tiny freezing probability of individual drops; likewise continuous flow IN detectors have too short a residence time to sample such a process. New observations are required to characterise this mechanism.

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