OBSERVATIONS OF THE ORIGIN AND DISTRIBUTION OF PRIMARY AND SECONDARY ICE IN CLOUDS

A THESIS SUBMITTED TO THE UNIVERSITY OF MANCHESTER FOR THE DEGREE OF DOCTOR OF PHILOSOPHY (PhD) IN THE FACULTY OF ENGINEERING AND PHYSICAL SCIENCES

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A detailed understanding of cloud microphysical processes is crucial for a large range of scientific disciplines that require knowledge of cloud particles for accurate climate and weather prediction. This thesis focuses on 3 measurement campaigns, encompassing both airborne and ground based measurements of the microphysical structures observed in cold, warm and occluded frontal systems around the United Kingdom, stratocumulus clouds in the Arctic and many different clouds observed over a 6 week period at a high-alpine site in the Swiss Alps. Particular attention was paid to the origin and distribution of both primary and secondary ice and the dominant features associated with ice phase processes.

During investigation of cold, warm and occluded frontal systems associated with mid-latitude cyclones around the U.K., secondary ice was often found to dominate the number and mass concentrations of ice particles in all systems. The presence of large liquid droplets was sometimes observed in close proximity to regions of secondary ice production. The existence of these provides a possible mechanism by which rime-splintering is greatly enhanced through the creation of instant rimers as the large drops freeze. In-situ measurements during the cold frontal case were used to calculate rates of diabatic heating during a comparison between bin-resolved and bulk microphysics schemes.

Observations in arctic stratocumulus clouds during spring and summer seasons revealed higher ice concentrations in the summer cases when compared to the spring season. This is attributed to secondary ice production actively enhancing ice concentrations in the summer, due to the higher temperature range the clouds spanned.

At Jungfraujoch in the Swiss Alps, ground based measurements allowed us to obtain high spatial scale resolution measurements of cloud microphysics and we found transitions between high and low ice mass fractions that took place on differing temporal scales spanning seconds to hours. During the campaign measurements of aerosol properties at an out of cloud site, Schilthorn, were made. When analysing a Saharan Dust Event that took place a possible link between the number of U.V. fluorescent particles and the number of ice particles was found in the temperature range around -10°C.
The University of Manchester

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CHAPTER ONE

INTRODUCTION

1.1 Motivation

Knowledge of the formation, distribution and influence of ice in the Earth's atmosphere and its role in cloud microphysics is of great importance to scientific disciplines focusing on vastly differing spatial and temporal scales. Since Benjamin Franklin suggested in 1784 that 'much of what falls as rain on the surface is likely to have started as ice' we have made great progress in our understanding of cloud microphysics. However, the measurement and representation of cloud particles in models still presents a significant challenge to those predicting global scale changes over centuries, as with General Circulation Models (GCMs), and those involved in the forecasting of synoptic and mesoscale meteorology over much smaller time periods. The lack of a thorough understanding of the ice phase in the atmosphere leads to motivation based solely on the natural instinct to simply understand the unknown, and a complete understanding is critical to predicting natural hazards that arise from a huge range of atmospheric processes, such as from mesoscale convective storms that produce severe weather, to the long term impacts of climatic change.

1.2 Cloud-Aerosol Interactions

The interaction of aerosol particles with clouds causes changes in their properties. The availability of Cloud Condensation Nuclei (CCN), along with the vertical wind velocity, determines cloud droplet number concentration, which influences droplet size, whilst the ability of some aerosol particles to act as Ice Nuclei (IN) has a significant impact on cloud processes (Chapter 2.1 describes the different pathways of ice nucleation in the atmosphere). The availability of CCN and IN can lead to changes in the amount (Ramanathan et al., 2001) and distribution of precipitation (Andreae and Rosenfeld, 2008), latent heating (Dearden et al., 2014), cloud radiative forcing and cloud lifetime. High concentrations of CCN lead to a tendency for smaller cloud droplets that reduce the
efficiency of collision coalescence, leading to suppression and delay of precipitation. Ice crystals, once formed, grow rapidly by the Bergeron-Findeisen (B-F) Process (Bergeron, 1937) and through riming in the presence of liquid water, releasing the latent heat of freezing. This process may have different impacts depending on the availability of IN and whether secondary ice production through the Hallett-Mossop (H-M) (Hallett and Mossop, 1974) (See Chapter 2.3) process is active. The development of relatively low concentrations of ice may lead to the growth of larger particles that rapidly cause the development of precipitation. If these particles fall through the H-M temperature zone then high concentrations of ice may be produced very quickly, leading to latent heating great enough to cause a buoyancy effect and influence the timing of any precipitation. A delay may lead to redistribution of precipitation as cloud system dynamics carry cloud water and precipitation away from its initial position. There is evidence that the generation of extra buoyancy, through latent heating as cloud water freezes, can impact on the development of convective cloud (e.g. Soong and Tao, 1980) and evidence recently suggests this can also have an impact on frontal clouds (Dearden et al., 2014).

### 1.3 Cloud-Radiative Interactions

The influence of microphysics on cloud radiative properties is complex and largely determined by the amount of condensed water, hydrometeor size, phase and habit (Curry et al., 1996) with these variables controlled by CCN and IN availability, together with secondary ice processes. Cloud albedo in predominantly liquid clouds is dependent on the liquid water path and the effective droplet radius. The first indirect effect (Twomey, 1974) predicts that an increase in CCN, in clouds with a constant liquid water path, will cause a decrease in effective droplet radius and an increase in cloud albedo (Twohy et al., 2013). The development of ice in clouds has a significant impact on liquid droplets as ice crystals grow rapidly, causing them to evaporate, which increases the optical depth of the cloud but reduces the albedo. As reported by Hogan and Illingworth (2003), a liquid supercooled layer often exists at cloud top when temperatures are higher than ~ -25 °C, acting to maintain a high cloud albedo. This is due to small ice crystals nucleating near cloud top falling out of this liquid layer and growing rapidly lower in cloud (Crosier et al., 2011). In very cold clouds e.g. cirrus, where large numbers of ice crystals scatter radiation at cloud top, then the complex shapes of the crystals play a key role in the scattering of light (Kaye et al., 2008; Takano and Liou, 1988; Takano and Liou, 1995).
1.4 Modelling

High resolution modelling of clouds and precipitation is important for research activities and the production of operational weather forecasts. The impact of aerosol particles on cloud microphysics is important to understand as this influences the ability of models to capture variability in cloud structures that may be caused through the presence of IN, for example. These processes are treated with varying levels of sophistication but remain a weak point in all models regardless of resolution (IPCC 5th assessment report, chapter 7, section 7.2.2.1). The treatment of the ice phase in research based models and operational models can differ significantly. The U.K. Meteorological Office Large Eddy Model (LEM) is a model for large-eddy simulation and cloud resolving modelling (CRM) (Gray et al., 2001). It represents homogeneous ice nucleation and individual modes of heterogeneous ice nucleation, for example deposition freezing (Meyers, DeMott and Cotton, 1992), contact nucleation (Young, 1974) and immersion freezing (Bigg 1953a). Also included is a treatment of secondary ice production through rime-splintering. The model is used to develop physical parameterizations for the Unified Model (UM), which contains a simple primary heterogeneous ice nucleation scheme (Fletcher, 1962). A major weakness of the UM used by the Met Office is the absence of any treatment of secondary ice production through the Hallett-Mossop (H-M) Process (Wilson and Ballard, 1999). This can lead to the model under estimating the glaciation of shallow slightly supercooled clouds that span the H-M temperature zone between -3°C < T < -8°C (Hogan et al., 2003). The development of CRMs (e.g. the Met Office LEM), which are generally considered to be models with sufficient resolution to explicitly simulate individual clouds and run for long enough to represent many different clouds and cloud lifetimes (Randall et al., 2003), has advanced our understanding of the relationship between cloud particles and cloud system properties. For example, comparison of in-situ and remote sensing studies with CRM simulations. As an example of this, Blossey et al. (2007) compared in-situ measurements made around the island of Kwajalein with results from a three dimensional CRM model finding under predictions of cloud amount and over predictions of radar reflectivity by the model. Dearden et al. (2012) performed simulations of a mixed-phase orographically induced wave cloud and compared them directly with observations from in-situ measurements made by aircraft and Connolly et al. (2013b) found evidence for an aerosol effect on tropical island thunderstorm Hector using a mesoscale CRM.
One of the key challenges faced by GCMs is the representation of variations in cloud microphysics on spatial scales much smaller than GCM grid boxes. Interactions between clouds and aerosol are sensitive to variations in turbulent motion, and while high resolution models can resolve these changes explicitly, providing greater information for determining cloud microphysical structure, GCMs do not have this capability. Larson and Golaz (2005) comment that forcing microphysics parameterizations with grid box averages of properties such as Liquid Water Content (LWC) results in inaccuracies, and models should determine some information about subgrid variability to use as input to any microphysics scheme. Subsequently development of parameterizations for use in GCMs to better resolve these small, subgrid scale variations statistically is underway, but these have not been implemented in any Coupled Models within the Intercomparison Project Phase 5 (CMIP5) simulations. Although problems of resolution have an impact on simulations, there is also a fundamental lack of understanding of ice and mixed phase clouds. The poor treatment of cloud microphysics in GCMs impacts on simulations of global circulations, such as the Hadley cell, and the distribution of precipitation and estimations of cloud adjustments due to microphysics interactions with aerosol particles (IPCC, 5th assessment report, chapter 7, section 7.2.2.2).

1.5 Microphysics Measurements

Accurate measurement of the size, shape and concentration of hydrometeors is fundamental to understanding cloud properties. The data presented in this thesis have been obtained using state of the art microphysics probes. Data analysis techniques made it possible to discriminate between liquid and ice particles in the studies presented, which is crucial for calculating statistics in relation to the ice phase. Each dataset was examined to identify and remove shattering artefacts that have plagued previous calculations of ice number concentrations. A description of the data analysis techniques used in this thesis can be found in Chapter 3.2.

1.6 Research Goals

In this thesis work from a number of different campaigns is presented. The motivation for each of these projects is outlined below.
1.6.1 DIAMET

The Diabatic Influence on Mesoscale Structures in Extratropical Storms project took place around the United Kingdom during 2011 and 2012. The overarching theme was to investigate the role of diabatic processes in generating mesoscale potential vorticity (PV) and moisture anomalies in cyclonic storms with a view to improving our ability to predict severe weather events. The main focus of work presented here is the measurement of the microphysics of cyclonic systems, in particular windstorms and intense precipitation events associated with mid-latitude cyclones, and the use of these measurements to calculate latent heating rates.

1.6.2 ACCACIA

The Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaign took place in the Arctic during spring and summer 2013. The project aimed to reduce uncertainty on the effects of aerosols and clouds on the Arctic surface energy balance and climate. The motivation for the work presented in this thesis was the measurement of arctic stratocumulus cloud microphysics and the comparison between previous observations and between cases within the Spring and Summer campaign windows. This will provide more detailed information on the physics of arctic cloud and will feed into better understanding of the role and persistence of these clouds.

1.6.3 INUPIAQ

The Ice Nucleation Process Investigation and Quantification project took place during January and February 2014 at Jungfraujoch, Switzerland. One of the key objectives of the project is to identify transitions between liquid and glaciated conditions that have previously been observed at Jungfraujoch, and to see if these are due to changes in aerosol concentration and composition. This will help us to understand the role of a range of aerosol types on the glaciation process in tropospheric clouds. Another key aspect is that because the instruments are located at ground sites the measurements of the microphysics can be made with very high spatial resolution. This will help us identify when clouds consist of neighbouring regions of largely glaciated cloud and cloud consisting of largely
supercooled water with little ice and when regions of cloud are truly mixed phase with ice and liquid water together in the same volume.

1.7 Thesis Overview

The main theme throughout this thesis is to make detailed measurements of cloud microphysics and analyse the dominant features within each case study. As discussed in Chapter 1 there is significant uncertainty surrounding the many different aspects of cloud microphysics, particularly the ice phase, that include the measurement and use in modelling studies.

This thesis is structured as follows. Chapter 2 describes the literature detailing the different pathways to ice formation in the atmosphere and the aerosol particles that are thought to influence it. Chapter 3 presents the methodology followed when carrying out data analysis, including the instruments used, measurement techniques and the analysis approach. Chapters 4 presents a paper that details microphysics measurements made in cold, warm and occluded frontal systems over the UK during the DIAMET campaign. Chapter 5 provides a summary of Dearden et al. (2014) paper entitled "Diabatic heating and cooling rates derived from in-situ microphysics measurements: A case study of a wintertime UK cold front". The work is focussed on the cold front described in Chapter 4 and calculated latent heating rates due to the various phase transitions associated with liquid water in the cloud system.

Chapter 6 is a paper that compares microphysics measurements in spring and summertime arctic stratocumulus during the ACCACIA campaign, focusing in particular on the origin of ice in these clouds and the impact secondary ice production has on cloud system structure. Chapter 7 is again a paper that details measurements made during a ground based campaign at high-alpine site Jungfraujoch as part of the INUPIAQ campaign. Swift transitions between ice and liquid are observed at this site and a potential link between biological aerosol and ice crystal number concentrations is investigated. Chapter 8 provides a summary of the main conclusions in this thesis together with a discussion about the implications for future work.
2.1 Ice Nucleation

In the atmosphere there are two distinct mechanisms by which water freezes to form ice crystals. The pathways are split into homogeneous and heterogeneous ice nucleation.

2.1.1 Homogeneous Ice Nucleation

The formation of ice crystals from liquid droplets in the absence of an aerosol particle acting as an Ice Nuclei (IN) is termed homogeneous ice nucleation. In the atmosphere this process takes place when the temperature is below ~ -35°C (Pruppacher and Klett, 1997) and involves the absence of impurities that initiate liquid to solid phase transitions taking place through heterogeneous ice nucleation. The process occurs as random fluctuations in the supercooled liquid molecules lead to embryos of the stable solid phase within the existing metastable phase. When a critical size of these molecular clusters is reached they can initiate the freezing of the rest of the liquid droplet. It is generally assumed that all droplets of pure water will freeze if the temperature is below -40 °C.

2.1.2 The Homogeneous Nucleation Rate

Classical Nucleation Theory (CNT) describes the stochastic process of homogeneous ice nucleation, which is described in Pruppacher and Klett (1997). The nucleation rate $J$ of pure supercooled water drops can be determined by the number of ice embryos that form per unit volume per unit time. It is generally thought that homogeneous ice nucleation is a stochastic process and therefore the homogeneous ice nucleation rate of a supercooled water droplet ($\omega$) is dependent on the number of molecules ($m$) in the drop and the probability ($\psi$) that a molecule will become the centre of a critical cluster during time interval $[0,t]$ (Riechers et al., 2013):
\[ \omega \equiv \frac{m \psi}{t} \tag{1} \]

Then the experimentally observed nucleation rate (\(\omega\)) is dependent on the volume (\(V\)) of the sample, the number of molecules (\(m\)) and the volume-dependent homogeneous ice nucleation rate coefficient \(J_v(T)V\):

\[ \omega = J_v(T)V \tag{2} \]

Table 1 shows how the nucleation rate (\(J\)) is a strong function of temperature, increasing rapidly with the degree of supercooling.

<table>
<thead>
<tr>
<th>Supercooling (°C)</th>
<th>(J) (cm(^{-3}) s(^{-1}))</th>
<th>Supercooling (°C)</th>
<th>(J) (cm(^{-3}) s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>29</td>
<td>(46 \times 10^{-11})</td>
<td>37</td>
<td>(30 \times 10^1)</td>
</tr>
<tr>
<td>30</td>
<td>(43 \times 10^8)</td>
<td>38</td>
<td>(50 \times 10^8)</td>
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<td>31</td>
<td>(15 \times 10^5)</td>
<td>39</td>
<td>(90 \times 10^9)</td>
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<td>32</td>
<td>(65 \times 10^{-3})</td>
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<td>33</td>
<td>(15 \times 10^0)</td>
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</tr>
<tr>
<td>36</td>
<td>(10 \times 10^6)</td>
<td>44</td>
<td>(20 \times 10^{17})</td>
</tr>
</tbody>
</table>

**Table 1.** The nucleation rate (\(J\)) as a function of temperature for homogeneous nucleation of ice in supercooled water, reproduced from Pruppacher and Klett (1997)

### 2.1.3 Heterogeneous Ice Nucleation

The presence of aerosol particles in the atmosphere can lead to the freezing of water from supercooled liquid droplets and directly from the vapour phase in ice supersaturated
This process is termed heterogeneous ice nucleation and is distinct from homogeneous ice nucleation due to the presence of an aerosol particle acting as an IN, triggering the transition from supercooled water to the solid ice phase. Although homogeneous ice nucleation is an important mechanism causing ice formation in temperatures below \(-35\) °C, at higher temperatures the presence of IN is required for primary ice formation.

### 2.1.4 Modes of Heterogeneous Ice Nucleation

There are four hypothesised modes of heterogeneous ice nucleation (Fig. 1). DeMott et al. (2003) describes these as i) Deposition freezing as ice embryos form directly on an IN from the vapour phase; ii) Immersion freezing caused by an IN that had previously become immersed within a supercooled liquid droplet (Bigg, 1953a,b); iii) Condensation freezing where a liquid droplet activates on a 'hybrid' aerosol particle that is acting as a CCN. Subsequently the droplet freezes as the particle also acts as an IN; iv) Contact freezing where a collision takes place between a supercooled liquid droplet and an IN causing the droplet to freeze.

![Figure 1: Schematic showing the modes of deposition, immersion, condensation and contact freezing (Adapted from Vali, 1999)](image_url)
There is still much uncertainty in the understanding of ice nucleation in the atmosphere, which Hegg and Baker (2009) highlighted by commenting that:

"It is impossible to observe the occurrence of nucleation directly in the atmosphere, but laboratory studies and numerical models suggest that the probability of occurrence of nucleation by any of these modes in a given atmospheric situation depends on environmental parameters (temperature, pressure, vapour partial pressure, vertical velocity), on the size, morphology and chemical nature of the catalyzing substrates, on the sizes of drops and on evaporation rates and even on air parcel history. As this list suggests, we know very little about the microphysics of heterogeneous freezing."

2.1.5 The Heterogeneous Ice Nucleation Rate

The heterogeneous formation of ice in the atmosphere is based on the characteristics of the foreign substrate coming into contact with a supercooled liquid droplet and on the time taken for an embryo of the stable phase to reach a critical size in which the phase change takes place. These two variables describe the composition and time dependent factors involved in ice nucleation. Vali (1994) commented that there is much debate over the relative importance of each process due to the way ice is modelled in clouds. There are two distinct descriptions of heterogeneous ice nucleation that either follow stochastic theory, in which given enough time, all droplets would eventually freeze provided supercooling is maintained, or singular theory, where a droplet freezing depends only on the minimum temperature reached, suggesting that aerosol particles acting as IN will cause the formation of ice at a specific degree of supercooling.

2.1.6 Stochastic Theory

Stochastic theory assumes uniformity across a droplet population and that any given droplet has a similar probability of freezing from a single ice nucleating species. The number of drops frozen per unit time is given by:
\[
\frac{dN_i}{dt} = N_d A J
\]  

(3)

Where \(dN_i\) is the number of droplets frozen, \(N_d\) is the number of droplets, \(A\) is droplet surface area and \(J\) is the stochastic nucleation rate for a given species of IN.

### 2.1.7 Singular Theory

The singular theory of ice nucleation assumes that the importance of time dependence on ice nucleation is insignificant in comparison to the influence an aerosol particles surface characteristics can have on the initiation of freezing. This assumption suggests that nucleation will take place at a specific site under a specific set of environmental conditions. This leads to a particular IN type becoming active at a characteristic temperature \(T_c\), above which ice nucleation will not occur (Murray et al., 2012). The fraction of droplets frozen in a population that contain the same surface area of aerosol particles \(\sigma\) within them can be expressed:

\[
\frac{n(T)}{N} = 1 - \exp(-n_s(T)\sigma)
\]

(4)

Where \(n_s(T)\) is the number of surface sites per unit area which become active on supercooling from 273 K to \(T\) (Murray et al., 2011; Connolly et al., 2009)

### 2.1.8 Modified Singular Theory

Vali (1994) examined the validity of the singular theory by repeatedly supercooling droplets and measuring their freezing temperatures. Although the same droplets generally froze within a few degrees when repeatedly cooled, differences in the freezing point of up to 5 °C were observed. This finding does not support the standard singular hypothesis, which suggests upon repeated cooling all drops should freeze at the same temperature. In addition when the cooling rate was increased by a factor of six there was a 0.4°C change in the freezing temperatures, a finding consistent with earlier work by Vali and Stansbury.
(1964) that found a 0.2°C alteration in the freezing temperature. Although the change is small this finding is thought to demonstrate the time dependent nature of ice nucleation (Murray et al. 2012). When accounting for the influence of the cooling rate on droplet freezing temperatures the fraction of drops frozen can be given as:

\[
\frac{n}{N} = 1 - \exp(-n_s(T_{min} - \alpha)\sigma)
\]  

Where the variable \(\alpha\) is the temperature offset from a freezing spectrum for a cooling rate of 1 K min\(^{-1}\) (Murray et al. 2011; Vali, 1994)

2.2 Aerosol Particles as Ice Nuclei

IN are usually water insoluble solid particles (Pruppacher and Klett, 1997). The main groups of aerosol implicated in atmospheric IN activity include mineral dusts, soot and bioaerosols such as bacteria, fungal spores, pollen and diatoms (Hoose and Möhler, 2012). Laboratory experiments investigating IN focus mainly on the IN activity of specific aerosol species, where temperature and the supersaturation ratio with respect to ice \(S_i\) is carefully controlled and the 'onset' conditions of nucleation (highest temperature and lowest supersaturation ratios for the formation of ice) are recorded. Hoose and Möhler (2012) provide a review of these experiments that shows the variability in IN capability that is determined by changes in aerosol species and size. Submicron mineral dusts tended to require higher \(S_i\) and lower temperatures to initiate ice formation (generally < -20 °C). Supermicron particles of the same species displayed IN activity at higher temperatures (onset ~ -10 °C) and lower \(S_i\). Observations of natural clouds have suggested that African dust influences glaciation of clouds, but there is disagreement about the mode of nucleation and temperature at which these Aerosol Particles (AP) become active (Sassen et al., 2003; DeMott et al., 2003 and Ansmann et al., 2008).

Results from experiments with soot particles showed little overlap between studies as onset thresholds were spread over a wide range of temperatures and \(S_i\) values. For example, Kireeva et al. (2009) found soot acted as an IN at \(T= ~ -10°C\) in the immersion mode, while during deposition mode experiments Möhler et al. (2005b) found soot to initiate freezing around -35°C. Choularton et al. (2008) reported that carbon rich aerosols had been
found in ice crystal residuals measured at Jungfraujoch, Switzerland (Cozic et al., 2008). Some biological aerosol particles consist of bacteria, some of which produce a protein that has been found to be IN active at slightly supercooled temperatures. Lindow (1983) reported this ability through investigation of frost damage to plants caused by bacteria. Due to this capability Pseudomonas Syringie is used in production of Snowmax, for use in the winter sports industry. Experiments with different types of pollens (Diehl et al., 2001; Diehl et al., 2002) such as pine, birch, oak and grass found them to be inefficient deposition nuclei but found that all pollen species did act as condensation freezing nuclei at relatively warm temperatures (~ -8 °C) with 50% efficiency of freezing reached between -12 and -18 °C. The same pollen species were also found to be efficient immersion and contact mode IN, and were active in temperatures as high as -5 °C. This work confirms the importance of biological aerosols, in this case pollen, as IN at relatively high temperatures. Murray et al. (2012) provided an overview of available quantitative data on different IN species immersed in supercooled water droplets and found soot and mineral dust to dominate primary heterogeneous ice nucleation below about -15 °C. The only material found to nucleate ice above this temperature was of biological origin. Recent work by Creamean et al. (2013) demonstrated the influence of biological particles in the natural environment that had been transported from desert regions of Africa and Asia and went on to affect the glaciation and precipitation of high altitude clouds over the Western United States.

The range of onset conditions of different aerosol species and indeed within the same species under laboratory conditions (Fig. 2) demonstrates the difficulty in characterising IN active APs in an experimental setting. Observing IN activity by specific aerosol species in the natural atmosphere presents a significant challenge.
2.3 Secondary Ice Production

Murgatroyd and Garrod (1960) discovered discrepancies between the number of ice nuclei and the concentration of ice crystals in clouds they studied. This finding suggested the existence of a mechanism leading to the observed 'ice enhancement' through the production of secondary ice. During the period before the 'breakthrough' Hallett and Mossop (1974) paper, that suggested a relationship between ice splinter production and riming, there were a number of theories about the origin of high concentrations of ice in clouds that would be expected to contain few natural IN. Koenig (1963) studied cumulonimbus clouds in Missouri and concluded that "clouds having large liquid water drops rapidly form high ice concentrations, regardless of the concentrations of foreign ice forming nuclei." These observations confirmed the discrepancy between IN concentrations and the number of observed ice crystals. Concentrations of ice that exceeded the expected IN concentration in cumulus clouds had been observed before and according to Koenig, attempts to explain this had rested on theories of mechanical fracture, detachment of fragile ice structures or
the shattering of drops as they freeze. Findeisen (1938) was the first to observe splintering of frost deposits (Koenig, 1963) and Kumm (1951) studied the mechanical breakdown of frost deposits and found that this required fragile ice structures and the temperature of the frost deposits needed to be altered constantly to cause splintering. This implied thermal shock may have been a mechanism of secondary ice production. Despite a possible link to fragile ice structures, Koenig (1963) didn't see any significant quantities of the fragile ice during investigation of the Missouri Cumulus that would possibly make ice crystals prone to breaking up, therefore it was thought to be unlikely that mechanical failure of ice particles could account for the high ice crystal concentrations observed. He outlined one mechanism, dependent upon thermal shock, by which ice crystal concentrations exceeding available ice forming particles might be achieved. It was argued that the freezing of large liquid drops that come into contact with ice particles could produce splinters through thermal shock of the parent ice crystal. Changes in temperature of the supercooled liquid water drop and the ice particle during the completion of freezing could create enough stress to cause the ice particle to fracture. However the large drops were few in number and it seemed unlikely this process could generate enough shattering events to account for the enhanced ice concentrations that were observed.

Mason and Maybank (1960) conducted laboratory experiments in which they observed suspended drops and nucleated them either heterogeneously or by contact with an ice crystal. They found drops in the range of 100 µm to 1 mm produced on average 20 to 50 splinters upon freezing. They observed drops freezing in a distinct pattern with an outer shell forming first followed by the solidification of the interior of the drop, the timing of this being dependent upon the ambient temperature. During this process cracks in the drops were observed along with spicules and seeping of water through the ice shell from the interior. Shattering and splinter production was not shown to be sensitive to drop size but dependent on temperature, becoming less efficient with decreasing temperature. The results presented by Mason and Maybank were unable to account for the multiplication rates observed by Koenig (1963). Mason and Maybank (1960) didn’t focus on any splintering from riming, but the idea was highlighted during work on thunderstorm electrification by Latham and Mason (1961). They conducted experimental work on charge generation by causing supercooled liquid drops to impact a hailstone target. They found that splinter production rates were dependant on temperature, velocity and drop size (particularly larger drops between 50-80 µm). For drop sizes < 30 µm splinter production declined. This early work was important in confirming the likelihood of a mechanism leading to the production of high concentrations of ice in cumulus clouds that far exceeded
the IN population. Droplet shattering, mechanical breakup and splinter production through riming were all implicated in the process with dependencies found on temperature, riming velocity and droplet size.

2.3.1 The Hallett-Mossop Process

The 'breakthrough' paper came in 1974 when Hallett and Mossop provided laboratory evidence that many ice splinters are produced during the riming process, now commonly referred to as the Hallett-Mossop (H-M) Process. They used a rod to simulate riming and it was moved in a circular path at up velocities up to 3 m s\(^{-1}\). When the rod was stationary a few crystals per minute were observed falling through a beam of light. When the rod was rotated there was an increase in the number of ice crystals observed that reached a constant after about 4 minutes. This first paper found a dependence on temperature, and subsequent research found several variables influenced the rate of production of these secondary ice splinters. These variables are described below.

2.3.2 Temperature

Hallett and Mossop (1974) found this phenomena only occurred in a narrow temperature range, almost exclusively between -3 °C and -8 °C, with a peak around -5 °C. This was the reason why Mossop, Brownscombe and Collins (1974) failed to see any significant splinters when they ran similar experiments but at a temperature of -9 °C, which was too low for the production of secondary ice splinters. Mossop and Hallett (1974) commented that during the accretion of 1 mg of rime several hundred daughter particles may be produced and if these grow and rime themselves then there would be no difficulty in producing the observed ice multiplication. For the first time they had observed secondary ice production rates that were explained the high concentrations seen in certain clouds. The experiments were conducted over several days and splinter production rates were averaged for each temperature range. There were anomalies with some of the experiments, with some producing several hundred more splinters than others conducted at the same temperature and velocity.

Mossop (1976) confirmed that maximum splinter production occurred around -5 °C and that the process was practically confined to between -3 °C and -8 °C. Heymsfield and Mossop (1984) looked at which variable was more important in the H-M process, the
temperature of the cloud or the temperature of the riming surface. They found that a particle of high fall velocity in a cloud of high Liquid Water Content (LWC) could produce secondary ice splinters considerably below -8 °C. They used a cloud chamber with riming rods moving at a velocity of 1.8 m s⁻¹ to simulate the terminal fall velocity of 1-2 mm graupel particles. In one experiment the rods were unheated and in another an internal heater was used to raise the surface temperature of the rods by 1°C. The effect of this was to alter the cloud temperature at which maximum splinter production takes place, by approximately 1 °C (Fig. 3).

Figure 3: Splinters produced per milligram of rime. 2 curves of the same experiment with the riming rods heated and unheated (Heymsfield and Mossop, 1984).

They calculated the change in rime surface temperature that would take place by altering graupel diameter and LWC and predicted the cloud temperatures that splinter production would take place at after these elevations in riming surface temperature. With low LWC
30

splinter production was confined between -2.5 °C and -7.5 °C even when graupel diameter was increased. When LWC and graupel diameter was increased the H-M zone shifts to lower cloud temperatures and Heymsfield commented that in extreme cases of large graupel diameter (0.5 cm) and LWC approaching 5 g m⁻³ the elevation in surface temperature is 6°C, with the cloud temperature range between -8 °C and -14 °C. The results strongly suggested the surface temperature of the rime is most important in the H-M Process. Research into the physical mechanism (Griggs and Choularton, 1983) suggested that the high temperature end of the H-M zone could shift to colder temperatures when the drop size distribution is altered, but that the low temperature cut-off was insensitive to these changes.

2.3.3 Droplet Size

Mossop and Hallet (1974) found a dependence of splinter production on drops ≥ 24 µm. In experiments using riming rods rotated around a circular path at temperatures of -5 °C they counted ice crystal fallout rates. They found that about 10 splinters per second are produced per drop ≥ 24 µm diameter per cm⁻³. The findings were presented in their paper (see Fig. 4). The association with larger drops was confirmed after splinter production was reduced by adding NaCl nuclei into the cloud to deliberately reduce drop sizes.
Mossop (1976) found the rate of splinter production at -5 °C was roughly 1 ice splinter produced for ever drop ≥ 24µm that is accreted.

Mossop (1978) used riming rods to simulate riming at a temperature of -5 °C whilst altering the drop size spectrum of the cloud. He found evidence that splinter production was not only dependent on larger drops, but also on smaller drops < 13 µm. The data presented (Fig. 5) showed that simply increasing the number of large drops would not always produce increased splintering rates, particularly if this was at the expense of smaller drops.

The implication of these findings is that a dependence solely on the larger end of the drop spectrum is inaccurate. Increasing the concentration of larger drops and neglecting the smaller drops, as in experiment H13, does not increase the number of splinters produced.
In Q15, where larger drops over ~ 25 µm are reduced, splinter production rates are low. Experiment Z14 appears to have the drop size spectrum most conducive to rime-splintering. This is likely to be due to larger drops being present in sufficient quantities, but not at the expense of the smaller drops < ~ 13 µm.

Mossop (1985) presented drop size data from natural cumulus clouds studied in Australia in which ice multiplication had been observed in an attempt to try and recreate a similar spectrum in laboratory experiments (Fig. 6).

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Figure 6: Number of large and small drops and the ratio (n_s/n_L) between them (Mossop, 1985)

The data show the number concentration of small drops and big drops, A condition shown to be associated with the most efficient splinter production. It also shows the ratio of small drops to big drops (n_s/n_l) in the cloud. Mossop states that due to the difference in the collection efficiency of graupel for both small and large droplets (less efficient for smaller drops) the ratio of drops accreted (N_S/N_L), were N is number of drops accreted, will be about a factor of 3 less than the ratio of drop concentrations (n_s/n_l), where n is the total droplet number concentration. This implies the N_S/N_L values applicable to these clouds were between ~ 0.02 and 2. Mossop looked at the number of crystals produced per large drop accreted ≥ 25µm as a function of N_S/N_L over a range of velocities (Fig. 7). The relationship found that reducing N_S/N_L decreased splinter production, presumably because the physical mechanism is dependent in some way on small drops ≤ 12µm.
2.3.4 Velocity

Mossop (1976) was the first to vary the velocity whilst conducting his experiments. Unfortunately there was too much scatter in the results to draw any conclusions about the association between splinter production rates and velocity. Mossop (1985) studied the influence of velocity over the range $0.55 \text{ m s}^{-1} - 5 \text{ m s}^{-1}$, a larger range than had been investigated before. Peak production of splinters was found at velocities of $1.8 \text{ m s}^{-1}$ and $3.6 \text{ m s}^{-1}$ (Fig 8), where splinter production was around 300 crystals per milligram of accreted rime. Either side of these velocities splinter production decreased.

Figure 7: Figure from Mossop (1985) showing the number of ice crystals per large drop $\geq 25 \mu\text{m} \ (A/N_L)$ accreted as a function of the ratio of small and large drops accreted ($\leq 12 \mu\text{m}) \ (N_S/N_L)$. Circles represent experiment means and vertical bars the probable error.
Saunders and Hosseini (2001) expanded the velocity range even further, studying splinter production over the range 1 m s\(^{-1}\) to 12 m s\(^{-1}\). The peak production was found at 6 m s\(^{-1}\) with 70 crystals produced for every milligram of rime accreted. These findings are in contrast to Mossop (1985) who found a peak of 300 crystals per milligram of rime, but at lower velocities.

### 2.3.5 Physical Mechanism

Mossop (1976) provided a summary of the different mechanisms that were proposed to be the cause of rapid glaciation in certain clouds. (1) The formation of an ice shell around the exterior of an accreted freezing drop that subsequently shatters as freezing advances. This has been observed in experiments on isolated drops (Mason and Maybank, 1960); (2) Drops that freeze after glancing a riming agent: It was suggested an ice dendrite could grow into a glancing drop and break off, being carried away with the cloud droplet, however glancing drops have been shown not to freeze on contact with a riming surface.
(Aufdermaur and Johnson, 1972); (3) The growth and detachment of fragile ice needles: Where ice growth is favoured from the vapour, needle shaped ice formation may grow from the rime. This seems plausible given the spike in splinter production at -5°C, a temperature at which the crystal habit promotes the growth of needles; (4) Detachment of ice by evaporation: Thin ice structures may be cut off in conditions where evaporation of the rime structure is taking place, however Mossop (1976) points out that this should mean increasing splinter productions at higher velocities, due to ventilation effects, something which hasn't been observed.

Mossop (1976) originally ruled out the idea of splintering due to the development of an ice shell as the symmetrical freezing pattern needed to produce one didn't seem likely during the growth of rime. However Choularton, Latham and Mason (1978) provided evidence to suggest that this mechanism, already documented for the freezing of isolated drops, could be implicated in secondary ice production through riming. Mossop (1978) showed that a mixture of small and large drops were needed to produce high splinter production rates. This configuration of the drop spectrum will produce riming events in which the larger drops freeze on contact with a small drop, creating poor thermal contact with the riming surface and fairly symmetrical heat loss to the environment, leading to the formation of an ice shell and protuberances. Mossop (1985) found reducing the ratio of small drops accreted to large drops accreted decreased the efficiency of splinter production, presumably because the larger drops then have less chance of contact with a small drop. Choularton et al. (1978) provided images of rimed ice crystals that they had grown and allowed to fall through a chamber cloud at -8 °C, riming as they did so before settling onto a formvar solution where a replica was created (Fig. 9).

![Figure 9: Rimed ice crystals displaying protuberances (Choularton et al., 1978)](image-url)
The image shows the rimed ice crystal replicas. The droplets have frozen, with little spreading on the rime surface, and interestingly the development of what appears to be a protuberance. This image suggests that drops freezing symmetrically, forming an ice shell and displaying protuberances could be implicated in splinter production through the H-M Process. One uncertainty with this experiment is that it was conducted at -8°C, this is at the low temperature cut off for splinter production proposed by Hallett and Mossop (1974). These findings support the association between riming, the presence of protuberances on fairly symmetrically frozen drops and splinter production. The conclusion of work by Mossop (1978) designed to examine the influence of the drop size distribution on splinter production was in contrast to Choularton et al. (1978). Mossop suggested that small drops play a role in producing a frail rime structure with numerous protuberances and the big drops interacting with this frail rime surface cause the splinters to be produced.

Mossop and Wishart (1978) 'revived' the drop shattering theory, finding increased splinter production at low velocities, over the range 1.4 to 3 m s⁻¹ and they suggested this was due to less spreading of drops upon impact with the riming rods and more symmetrical freezing which would promote an ice shell and splinter ejection. It's not clear why they observed higher rates of splinter production at lower velocities and this result is in contrast to work by Mossop (1976) who found evidence for decreasing splinter production rates at lower velocities. An extensive study of velocity by (Saunders and Hosseni, 2001) found a peak in splinter production at higher velocities and an increase in splinter production over the range 1.4 to 3 m s⁻¹. Mossop (1980) provided more support for the drop shattering theory proposed by Choularton et al. (1978). Drops of distilled water were frozen at -8°C, one sample was contaminated with Ammonia (NH₃), the desired result of adding NH₃ was to weaken the ice structure. Roughly half of the unpolluted sample developed protuberances whilst the formation was reduced in the drops doped with NH₃. They tested the sensitivity of splinter production to the introduction of NH₃ into a chamber cloud whilst riming rods were simulating riming. When NH₃ was introduced they found a reduction in splinter production to 9% of levels observed with an unpolluted cloud. This finding provides strong support for the ice shell theory because by altering the cloud chemistry to deliberately inhibit ice shell and protuberance formation a subsequent reduction in splinter production was observed.

Choularton et al. (1980) studied protuberance formation over the temperature range -1 °C < T < -12 °C and over riming velocities 1.5 m s⁻¹ to 3.0 m s⁻¹. Protuberance production was
found to be greatest around -5 °C, about 1 for every 20 drops of diameter > 20μm. Protuberances were restricted to the temperature zone identified through work by Hallett and Mossop, between -3 °C and -8 °C. At higher temperatures the cut off seemed to be associated with the spreading of drops due to a slower rate of freezing, reducing the chance of symmetrical freezing. At temperatures below -8 °C many symmetrically frozen drops were observed, but no protuberances detected over the velocity range 1.5 to 3.0 m s\(^{-1}\). The image (Fig. 10) of a droplet 35 μm in diameter was collected at a velocity of 1.5 m s\(^{-1}\) at a temperature of -7 °C.

![Figure 10: Frozen droplet displaying protuberance and possible ejected ice splinters (Choularton 1980)](C)

This image shows a symmetrically frozen drop displaying a protuberance and what seems to be a disturbance above this. Unfortunately it’s not clear from the image what the exact nature of this disruption is, but it is possible that a thin jet of water has been ejected from within the freezing interior of the drop through the protuberance, causing part of the ice shell to shatter and eject splinters. Some of these would have been ejected into the chest freezer during riming and a small number could have been left behind attached to the jet of water that froze during ejection from the droplet. Choularton et al. (1978) suggested that the presence of protuberances was indicative of the formation of an ice shell, but this image, together with findings that protuberance production as a function of temperature, riming velocity and drop size distribution is paralleled strongly with splinter production, suggests that protuberances are directly involved in the ejection of secondary ice splinters.
Griggs and Choularton (1983) investigated protuberance formation through rimeing when using a combination of large and small drops, only large drops and only small drops. They found formation occurred with a combination of large and small drops and also that with just small drops. The presence of protuberances was observed throughout the H-M temperature zone. When they used only large drops the temperature range was reduced (-4.5 to -9 ºC). The finding that rime-splintering was active in the presence of small drops only is in contrast to research by Mossop (1985), that found splinter production was dependent on the presence of both small and large liquid droplets. It is possible that in the study by Griggs and Choularton (1983), larger drops ≥ 24µm were present in small quantities, providing the conditions needed for symmetrical freezing. As well as investigating the role of drop size on splinter production, they looked at the freezing patterns of drops in carbon tetrachloride or freon and silicone oil to find an explanation for these observations. They outlined 3 distinct modes of freezing associated with the nucleation of drops and the formation of an ice shell. 1) At temperatures above -5 ºC an ice front grows outwards from the rimer with ice formation enhanced around the edges of the drop; 2) At temperatures below around -5 ºC there are two ice fronts, one from the riming substrate and another from the opposite edge of the drop, with enhanced ice growth rate around the drop edges; 3) At temperature below -9 ºC, freezing is seen to be 'symmetrically inwards'. The two modes of freezing at higher temperatures have the potential to produce protuberances, whilst temperatures below around -8 ºC will not freeze in a way which promotes a thick ice shell and hence little protuberance occurrence. These observations provide an explanation for the low and high temperature cut off for splinter production. Below -8 ºC the increased rate of heat loss to the environment allows the formation of a stronger ice shell relative to those formed at higher temperatures. The increased pressure during the completion of freezing of the interior of the drop is not sufficient enough to rupture this stronger ice shell and cause protuberance formation. At higher temperatures drops are likely to spread before being able to freeze symmetrically. These patterns are caused by the extent of the first stage of freezing as the rimed drop warms to 0 ºC.

Dong and Hallet (1989) Grew rime in a wind tunnel over the velocity range 0.2 m s⁻¹ to 2 m s⁻¹ and a temperature range of -1 ºC to -13 ºC. They failed to see evidence for the symmetrical freezing that is proposed to lead to ice splintering. This led them to propose a model of thermal shock to explain breakup of ice crystals. Previous work by King and Fletcher (1976) considered a model of thermal shock that led to a negative result. They experimented with raising the temperature of an ice crystal by causing it to come into

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contact with water. A portion of the ice crystal surface would rise in temperature rapidly to 0 °C. The median temperature at which the ice crystals would crack was -16 °C. Dong and Hallett proposed a different method of thermal shock breakup (Fig. 11).

Figure 11: Schematic of splintering through a method of thermal shock (Dong and Hallet, 1989)

It is assumed the temperature of the droplets, ice particle and the ambient air are the same. As the drop contacts the ice surface latent heat raises the surface of the drop to 0 °C. Freezing completes and the surface temperature falls, leading to the fracturing of the frozen drop. These findings are in contradiction to the association of protuberances with ice splinters and evidence to show that their deliberate suppression through introduction of NH₃ reduces splinter production (Mossop, 1980). Overall the balance of evidence favours the Griggs and Choularton mechanism of shattering ice shells as the cause of the secondary ice particle production by the H-M mechanism.

### 2.3.6 Ice Enhancement in Natural Clouds

Hobbs et al. (1980) studied cumulus clouds in Montana and calculated ice enhancement ratios (Fig. 12), defined as the maximum concentration of ice particles measured in a cloud to the concentration of ice nuclei at cloud top temperature.
Figure 12: Ice enhancement ratios as a function of cloud top temperature (Hobbs et al., 1980)

In the Hallett-Mossop temperature zone there were significant discrepancies between IN and ice crystal concentrations. In addition to this there is evidence for enhancement in the temperature zone -12 °C to -15 °C. In this temperature regime ice crystals follow a dendritic growth habit, these characteristic ice particles are fragile crystals that some have suggested (Koenig, 1963) could fracture to provide a source of secondary ice. Hobbs and Rangno (1985) studied the development of ice in 90 cumuliform clouds of maritime and continental origin. They found 35% of the clouds they studied did not meet the criteria for the H-M Process to operate. Where strong ice enhancement was observed in stratiform clouds there were insufficient concentrations of small drops < 13µm and most of these clouds did not contain graupel. It was also seen that maritime clouds didn't contain sufficient concentrations of smaller drops. Hobbs and Rangno claim that these findings suggest the H-M Process cannot explain all ice enhancement and that although capable of explaining some of the ice multiplication in clouds is actually secondary to a more powerful mechanism of ice enhancement. They proposed that contact nucleation at the top of the clouds studied could cause ice multiplication. They argue that mixing of ambient air into the cloud will encourage the evaporation of some of the cloud drops. At this point thermophoretic forces will cause aerosol to move towards the evaporating drops, coming into contact with and potentially causing ice nucleation. Hobbs and Rangno point out that if there are submicron aerosols in concentrations of $10^6$ L$^{-1}$ even in maritime clouds, then
about 1 in every $10^4$ would need to come into contact with cloud droplets to produce ice concentrations of several hundred per litre.

Hobbs and Rangno (1990) found maritime cumulus clouds contained high concentrations of ice that in one case they believe developed too quickly ($\leq 1$ to 1100 L$^{-1}$ in 12 minutes) to be explained by H-M splintering. They suggested rapid glaciation of the cloud through the activation of IN in regions of localised high supersaturations with respect to water. They suggested larger drops sweep up many of the smaller drops, a sink for water vapour, creating conditions for high supersaturations of 5-10%. Under these conditions deposition or condensation freezing may become active due to high supersaturations with respect to ice, producing an effective ice enhancement mechanism. The concentrations observed by Hobbs and Rangno of 1100 L$^{-1}$ in the cumulus they studied were too high to be explained by the process of contact nucleation suggested by Rangno and Hobbs (1985).

Hobbs and Rangno (1991) Calculated that ice concentrations in small polar maritime cumuliform clouds that they studied were an order of magnitude higher than could be explained by the H-M splintering process. They explain the appearance of frozen drops shortly before large quantities of ice crystals in terms of two separate nucleation processes. The first is a thermophoretic one encouraging the migration of IN towards liquid drops after ambient air is mixed into cloud tops, triggering the appearance of frozen drops. The second process is activation of IN by high supersaturations and is said to be the cause of sudden appearance of the high concentrations of ice crystals. Although these two processes, both encouraging ice nucleation, may occur at similar times it's unclear how they could bring about a coordinated development of frozen drops, quickly followed by high concentrations of ice crystals. The findings that frozen drops and graupel are present shortly before ice multiplication takes place is more likely to be explained by those initial frozen ice particles riming and producing secondary ice splinters through the H-M process.

Rangno and Hobbs (1994) studied continental cumuli clouds finding ice concentrations were correlated with the broadness of the droplet spectrum at the top of the cloud. They concluded that contact nucleation through the thermophoretic force created by the evaporation of cloud drops cannot explain the high concentrations of ice observed in these clouds, but still believe the H-M splintering process, in its current form, cannot account for the ice enhancement. They concluded by stating that "in summary, the origin of ice in cumuliform clouds remains a mystery".
Blyth and Latham (1997) use a multi-thermal model of cloud glaciation by the H-M process. Blyth and Latham (1998) commented on numerous papers by Hobbs and Rangno that claimed the H-M process couldn't account for ice multiplication. They outlined that their model tracked the trajectories and life histories of all graupel and ice crystals as they moved around the cloud, grew and produced their own splinters. They applied their model to the cumulus cloud studied by Rangno and Hobbs (1991) and were able to produce the required multiplication rates to explain the observations in this cumulus cloud. They confirm the 2 stages of the process by which there is an initial low concentration of ice crystals followed by a very rapid increase in crystals at cloud top. Bower et al. (1996) observed a spike in ice crystal concentrations around -15 °C and highlighted the possibility of another ice multiplication process. However Mason (1998) suggested the crystals are formed in the H-M zone and carried in weak updraughts through the -8 to -12 °C level, growing slowly, before reaching the -15 °C temperature region, where they grow rapidly through vapour diffusion.

It is now known that previous measurements in natural clouds such as this were contaminated with shattered artefacts from ice crystals breaking up on probe inlets. The enhancement in ice crystal number concentrations, around -15 °C, falls within a temperature range known to produce fragile dendritic ice that leads to artificial enhancement of ice concentrations as Optical Array Probes (OAPs) ingest these ice crystals through their inlets. A discussion about these artefacts and their treatment in the dataset can be found in Chapter 3.2.3.

Despite this and the dominance of the H-M process when the cloud converges the appropriate temperature range there is discussion in the literature suggesting that other methods of ice multiplication may be active in some clouds. There is some evidence (see for example Pruppacher and Klett, 1997) that ice crystal multiplication can also occur by the shattering of liquid drops as they freeze and by the fragmentation of ice particles either on collision with other particles or as they evaporate. The proposed mechanism for the production of splinters on the freezing of a supercooled drop is that in the first stage of freezing ice is produced in the body of the droplet as it warms up to 0 °C. This then nucleates an ice shell which spreads round the drop as heat is lost to the environment. As the drop freezes further with a shell of ice the pressure in the drop rises and disruption of the shell occurs producing protuberances, the ejection of gas bubbles, supercooled water and ice fragments. It has been observed in laboratory studies that the drop may fragment into 2 or more parts on freezing. The contribution of this process to ice multiplication in
clouds is poorly quantified, however there is little evidence to suggest that this is a major source of secondary ice. For example Rangno (2008) found evidence that this process operated in shallow marine frontal clouds producing a modest number of ice particles consistent with laboratory studies but the process is not sufficiently powerful to explain a large enhancement of ice crystal concentrations such as is often observed.

Another process often reported in the literature is the fragmentation of ice crystals particularly delicate shapes such as stellar or dendritic crystals, see for example Griggs and Choularton (1986). There is laboratory evidence that this process occurs and some evidence of broken fragments in natural clouds. Many of the early studies that reported evidence of fragments around -15 ºC e.g. Bower et al (1996) were possibly contaminated by the fragmentation of ice crystals on the inlets of airborne probes as discussed above. Recent airborne studies in natural clouds have found little evidence for this process e.g. Lloyd et al. (2014a,b) and Crosier et al. (2011) in layer clouds and Crawford et al. (2012) in convective clouds. However Yano and Phillips (2011) have published calculations to suggest that this process may be important in clouds where rime-splintering by the Hallett-Mossop process is not significant.

A further discussion of the roles of secondary ice particle production by the H-M process and the possible roles of the shattering of frail ice particles and the fragmentation of freezing drops will be presented later in this thesis in the light of the data presented.
CHAPTER THREE

METHODS AND TOOLS

3.1 Methodology

The work presented in this thesis was developed from three field campaigns that took place in the Arctic, The United Kingdom and Jungfraujoch in Switzerland. These involved the measurement of cloud microphysical properties during airborne and ground-based detachments. The instruments and techniques used to analyse the data they produced are described in this chapter.

3.1.1 BAe 146-301

During the DIAMET and ACCACIA campaigns in-situ measurements were made with the use of the Facility for Airborne Atmospheric Measurements (FAAM) British Aerospace 146-301 (BAe 146-301) aircraft. This modified large Atmospheric Research Aircraft (ARA) has a range of ~ 1800 nautical miles and can climb to 35,000 ft, allowing measurements to be made in a wide range of meteorological conditions at different altitudes. The cruising speed of the aircraft is up to 796 km hr⁻¹, with a science speed of about 100 m s⁻¹ and flight duration of around 5 hours. The instrumentation used on the aircraft to gather the majority of the data used in this thesis will now be described.

The 3V-CPI instrument was deployed during a ground based campaign at Jungfraujoch only. Many of the remaining cloud physics probes were utilised at both Jungfraujoch and during airborne measurement campaigns. A complete list and description of all instruments operating on the aircraft can be obtained through FAAM (available online: http://www.faam.ac.uk).
3.1.2 Temperature Sensors

On board the BAe-146 deiced and non deiced Rosemount/Goodrich type 102 temperature sensors were used to obtain information about the ambient air temperature during science flights. Both probes have inlets designed to minimise water and particle ingress that could cause wetting of the sensor and temperature anomalies (Blyth, Cooper and Jenson, 1988), and the de-iced temperature sensor contains a heater to prevent the probe from icing up during flight. Most of the work presented in this thesis uses the de-iced temperature sensor.

3.1.3 Cloud Droplet Probe (CDP)

The CDP, developed by Droplet Measurement Technologies (DMT), Boulder, USA (Lance et al., 2010) (Fig. 13) detects liquid cloud droplets over the size range 3-50 µm. From these measurements information about droplet size and number concentrations can be calculated. In this case particle data was provided at 1 Hz and separated into 30 size bins for analysis. The width of the size bins is 1 µm between 3 - 14 µm and then 2 µm for sizes up to 50 µm. The CDP uses an elliptical gaussian beam (~ 2 mm x 0.2 mm) generated by a diode laser to measure individual particles and determine their size. Light scattered over the angles ~ 4-12 ° in the forward direction is collected by photo detectors that determine the particle size based on Mie scattering solutions. One potential source of error from this probe is the detection of ice particles that scatter light as the pass through the laser beam. The impact of this is poorly quantified, but the number of ice particles counted by this probe is likely to be limited to a few per litre.
3.1.4 Cloud Imaging Probes 15 and 100 (CIP-15 and CIP-100)

The CIP-15, and CIP-100 (Baumgardner et al. 2001) are optical array probes consisting of 64 element photodiode arrays with element resolutions of 15 and 100 µm respectively. The CIP-15 and CIP-100 measure particles over the size range 15-930 µm and 100-6200 µm respectively. Both of these probes operate on principles of shadow imaging to detect and create images of particles that pass through the probes sample volume (Fig. 14). The optical arrays in each probe are illuminated by a collimated laser beam from a 45-mW 0.685 µm wavelength diode laser. Each time the array moves a distance of 15 and 100 µm respectively (determined by the airspeed) the on/off state of each diode in the array is recorded. The CIP-15 is a greyscale probe in which diodes record a particle event at 25, 50 and 75% shadowing respectively. The CIP-100 is a monoscale probe that produces particle images based on 50% reduction in illumination of array diodes by the laser. The particle images provided allow analysis software to calculate statistics about individual particle shape and size (see Ch. 3.2)
3.1.5 The Two-Dimensional Stereoscopic (2D-S) Probe

The 2D-S probe is another monoscale shadow imaging probe based on the same principles as the CIP-15 and CIP-100 that measures particles over the size range 10-1280 µm. The main feature that separates this probe is the presence of two photodiode arrays with overlapping laser beams. They record single images of particles outside of the overlap region and two independent images of the same particle where the beams overlap (Lawson et al., 2006). The probe also has a higher resolution (10 µm) and faster response (> 10 times faster) than the CIP probes. Lawson et al. (2006) describes the probe optical system, which is based on Keplerian telescope design. This produces a theoretical 5X magnification of particle shadow events onto the photodiode arrays, which each consist of 128 elements of 42.5 x 50 µm pixels (Fig. 15). This produces an effective imaged pixel size of 10 µm and maximum effective array dimension of 1280 µm. The two photodiode arrays are illuminated by single mode, temperature stabalised, fibre decoupled diode lasers operating at 45 mW. The array elements record a particle event at a 50% shadowing threshold.
3.1.6 Cloud Aerosol Spectrometer (CAS)

Measurements of aerosol during some of the work presented in this thesis were made with the use of the CAS probe. The probe uses light scattered from a 45mW gaussian mode diode laser with a wavelength of 0.685 µm in the backwards and forward directions (Fig. 16) over the 4-13°. The size of each particle is determined using Mie scattering theory and by assuming spherical particles of a known refractive index to produce an equivalent optical diameter (Baumgardener et al., 2001). The CAS measured particles in the range 0.51 µm to 50 µm. In addition to the forward and backward scatter detectors there is a backward s-polarized scatter detector, which gives the instrument the ability to detect the portion of light that has become depolarised by the scattering medium. We do not present any information about the depolarisation caused by cloud particles.
3.1.7 Passive Cavity Aerosol Spectrometer Probe (PCASP)

The PCASP-100X (Fig. 17) is an optical particle counter (OPC) that was used to provide aerosol measurements over the range 0.1 \( \mu m \) and 0.3 \( \mu m \). Particles that pass through the HeNe laser (wavelength 0.63 \( \mu m \)) scatter light, the intensity of which is measured over the 35-120° range. Particle size is determined using Mie scattering theory and classified into one of 32 size channels.

![Figure 17: The PCASP instrument design (DMT PCASP-100X Manual) showing how the instrument detects aerosol particles passing through the laser beam.](image)

3.1.8 Three-View Cloud Particle Imager (3V-CPI)

The 3V-CPI consists of a 2D-S shadow imaging probe and a Cloud Particle Imager (CPI) probe (Fig. 18) The operation of the 2D-S portion of this probe is identical to the standalone instrument that has already been described. The CPI part of the probe consists of a 1024 x 1024 CMOS camera that produces 2.3 \( \mu m \) pixel-size digital images of cloud particles. The camera of the CPI is triggered as particles are detected by the 2D-S particle detection system, which as a result triggers the CMOS camera that takes images of the particles passing through the probe sample volume. The probe produces images of liquid droplets and ice crystals over the size range 2.3 - 2500 \( \mu m \).
Figure 18: The PDS systems (Stratton Park Engineering Company) from the 2D-S portion of the probe, which trigger the CMOS camera as particles pass through the imaging laser.

3.1.9 Wideband Integrated Bioaerosol Sensor (WIBS)

Single particle UV-fluorescence measurements were made at the Schilthorn site (2,970 m asl) performed using a WIBS described by (Kaye et al., 2005; Foot et al., 2008; Gabey et al., 2010; Stanley et al., 2011). The WIBS used here was WIBS Model 4 and is described by Robinson et al. (2012). The single-particle elastic scattering intensity is measured in the forward direction at an angle of 90° and is used to calculate the particle equivalent optical diameter through Mie scattering solutions for spherical particles. The measurement range for this instrument is $0.5 < Do < 20 \mu m$, with 50% sampling efficiency at 0.8 µm (Gabey et al. 2010). The sizing measurement is used to trigger broadband UV pulses from xenon flash-lamps filtered at 280 nm and 370 nm, designed to excite biomarker molecules including tryptophan and nicotinamide adenine dinucleotide phosphate (NAD(P)H) respectively within a particle. Fluorescence resulting from this process is then measured over two selected waveband regimes.

3.1.10 Instrument Calibration

Calibration of instrumentation was carried out during each measurement campaign described in this thesis. The University of Manchester, Centre for Atmospheric Science CIP-15 and FAAM CIP-15 and 100 models were calibrated before and after each campaign they were involved in using a spinning disc containing targets in the size range
50 um < $D_p$ < 2000 um. During campaigns calibrations are carried out if there are problems with the instruments. Latex Spheres of known size are used in the calibration of the CDP, PCASP and CAS. During airborne campaigns these instruments are calibrated before each flight. Calibrations of the CDP and CAS during ground based measurement periods at Jungfraujoch took place during suitable times in moderate weather conditions.

3.2 Data Analysis Techniques

The methods employed to calculate the data products used in this thesis are presented below.

3.2.1 Cloud Particle Selection

During airborne campaigns the 2D-S, CIP-15 and CIP-100 OAPs were used to calculate concentrations of particles over the combined size range 10-6250 µm through examination of the shadow images produced by each probe. The 2D-S portion of the 3V-CPI was used in the case of the Jungfraujoch ground based deployment only. The principles of operation for all OAPs are the same in each project and so no changes were made to the way the data were analysed. These shadow imaging probes provide images of hydrometeors at varying pixel resolutions. Particles measured by the probes can be classified in three groups based on their position on the array as they pass through the probes sampling area. Fig. 19 shows examples of images from the 2D-S OAP that are entirely within the sample volume, obscuring one end element and obscuring both end elements. It has been found that a significant fraction of particles fall only partially within the sampling area of the probe (one or both end elements obscured) (Heymsfield and Parrish, 1978; Knollenberg, 1970).
Selecting particles that only fall entirely within the probe sample area reduces the number of particles a cloud imaging probe can measure and is particularly sensitive to increasing particle size. Due to this problem we have used the 'centre in' technique in work presented here, which increases the sample volume in the larger size ranges. A description of this sampling technique can be found in Heymsfield and Parrish (1976). Given any of the three particle orientations in Fig. 19 it is possible to determine the size of the image either directly (Fig. 19a) or through calculation of the particle size based on the portion of the particle that falls within the array. This is possible if the ratio of the dimension along the y axis to the dimension along the x axis of the partially imaged particle is ≥ 0.2. With this approach it is possible to determine the size of partially imaged hydrometeors using geometric calculations. Criteria based on the characteristics of each image event are used to filter particles that are likely to lead to artefacts in the data as a result of out of focus or shattered particles that have been detected. An image event is defined by a continuous extinction in each image slice of one or more pixels on the photodiode array. During processing an image event was accepted based on the number of individual particles in one image event and the fraction of the area of the largest particle in the image compared with the combined area of all other particles. The minimum and maximum area fractions were set to 0.8 and 1 respectively and the maximum number of particles accepted in an image event was 10. The maximum area fraction ensures that the largest particle in the image event is ≥ 80 % of the total area of other pixels in the image event. The criteria that an image event must have ≤ 10 individual particles reduced the chance of a shattered particles being analysed and contributing to the dataset. Examples of images rejected based on the criteria used are shown in Fig. 20.
Figure 20: Particle imagery from the 2D-S showing an image event that has been rejected due to an image fraction < 0.8 (a), a particle that has an image fraction > 0.8 but has been rejected due to number of individual particles in the event being > 10 (b), an image that has been rejected due to both the image fraction being < 0.8 and the number of particles being > 10. Image (d) shows a particle that has been accepted with an image fraction > 0.8 and number of particles ≤ 10.

3.2.2 Particle Size

When a particle passed the set acceptance criteria the size of the particles ($D_p$) was calculated by taking the mean of the maximum dimension along ($D_y$) and across ($D_x$) the probe array. This approach to sizing the measured particles is less sensitive to the presence of elongated particles than using just the maximum dimension in the across array direction $D_x$ and allows more consistent representation of particle size distributions in regions the dominant growth habit produces elongated ice particles, such as columnar and needle type crystals (Crosier et al. 2013). The use of particle maximum dimensions is also an approach used to determine particle size. While we the mean of $D_y$ and $D_x$ in this work, the use of maximum dimension of a particle is often used in modelling studies.

3.2.3 Filtering by Particle Inter-Arrival Time

Cloud microphysics probes are vital for studying cloud properties but it has been known for some time that cloud microphysics data, particularly ice number concentrations and size distributions, can be altered by breakup of natural ice particles on probe housing (Field et al., 2006). The two mechanisms that produce these artefacts are 1) Mechanical breakup through impact with the probe arms and 2) fragmentation due to the turbulence and windshear produced by probe housing (Fig. 21) (Korolev and Isaac, 2005).
During all airborne campaigns described in this thesis the 2D-S, CIP-15 and CIP-100 probes were fitted with anti-shatter tips (Korolev, 2011) to reduce the likelihood of ice crystals breaking up on probe housing. During the ground based campaign at Jungfraujoch the 3V-CPI was fitted with a 'knife-edge' inlet that was also designed to reduce shattering artefacts. Despite these modifications shattering is still likely to contribute to microphysics datasets (Crosier et al. 2013; Field et al. 2006). To remove these remaining particles Inter-Arrival Time (IAT) filtering was used to discount particles with unusually low IATs. When measuring cloud particles the 2D-S and CIP instruments provide particle time stamps that allow examination of particle IAT histograms. When shattering is present in the dataset these plots exhibit a bi-modal distribution, with a mode of 'good' particles centred to the right of a mode of particles with shorter IAT times that are assumed to be artefacts from shattered ice particles. The magnitude of the influence on measurements from shattered particles is variable depending on the microphysical conditions at the time any data is collected. Jensen et al. (2009) found that when natural ice crystal concentrations were low shattering artefacts enhanced concentrations by an order of magnitude. When high concentrations were observed the influence from a shattering mode was negligible. Crosier et al. (2013) presented IAT data from different temperature regimes that demonstrated the contribution from a shattering mode to data that was related to temperature, natural ice
crystal number concentrations and the size of the ice particles (Fig. 22). These plots show IAT histograms for different temperature regimes, with mean particle size in each IAT bin to provide information about the particle sizes in each IAT bin, and the cumulative distribution function (CDF) to show the contribution of each mode to the total number concentration. In plots a-c the mode centred around $2 \times 10^{-3}$ secs represents intact particles. In these plots the mode centred to the left of $1 \times 10^{-5}$ secs represents shattering artefacts. The mode of good particles doesn’t extend generally below $1 \times 10^{-4}$ secs and so selecting this value as a minimum IAT threshold is appropriate in these regions. The contribution from this mode of shattered particles can be significant (> 80% in (b)) and must be removed to prevent contamination of the dataset. In (d) natural ice number concentrations are enhanced through secondary ice production, shifting the mode of ‘good’ particles to be centred $\sim 2 \times 10^{-4}$ secs. In this case the IAT threshold of $1 \times 10^{-4}$ secs leads to the removal of natural ice particles and a threshold IAT of $1 \times 10^{-5}$ secs is more suitable for this region.

Figure 22: Figure from Crosier et al. (2013) showing particle inter-arrival time histograms (black, right axis) for the CIP-15 at (a) $-42^\circ$C, (b) $-18^\circ$C and from (c) within and (d) outside the NCFR regions at $-10^\circ$C. Also shown are the mean area equivalent diameter (blue) and the cumulative distribution function (CDF) of the number concentration (red).

During data analysis IAT histograms were analysed to identify the position and existence of any shattering mode and set an appropriate IAT threshold to remove these artefacts.
3.2.4 Phase Identification

Key to the work presented here is the identification of ice particle number concentrations in each of the measurement campaigns. Calculation of ice crystal number concentrations, mass loadings and size distributions was done through examination of the raw images produced by the Optical Array Probes (OAPs). The particle statistics used to discriminate between liquid and ice are the total area of the particle and the perimeter around the edge of the particle (Crosier et al., 2011). These parameters allow analysis of a particles circularity (C), with spherical particles assumed to be liquid droplets and assymetrical particles judged to be ice crystals. Eq. 6 shows the definition of circularity (C), where P is the particle perimeter and A the measured particle area. Particles with a circularity greater than 1.2 were classified as ice in the work presented. Only particles consisting of ≥ 20 pixels are used in phase identification. This is because particles containing fewer pixels lack the resolution needed to determine their shape.

\[ C = \frac{p^2}{4 \pi A} \] (6)

From separation of raw images into ice and liquid particles calculation of ice number concentrations and Ice Water Contents (IWC) were possible. For IWC calculation the mean particle size was determined and used the mass dimensional relationship of Brown and Francis (1995). The IWC calculation used for particles > 100 µm is shown in Eq. 7, where \( M \) is the particle mass in grams (g), \( D \) relates to the diameter in microns (µm), a = \( 7.38 \times 10^{-11} \) and b = 1.9. For particles below 100 µm this would imply a mass greater than a solid ice sphere. Any particle with diameter smaller than 100 µm is assumed to be a solid ice sphere, where a = \( 4.82 \times 10^{-13} \) and b = 3.

\[ M(D) = aD^b \] (7)

IWC is then,

\[ IWC = \sum_{D_{min}}^{D_{max}} n(D)M(D) \] (8)
CHAPTER FOUR

OBSERVATIONS OF THE ORIGIN AND DISTRIBUTION OF ICE IN COLD, WARM AND OCCLUDED FRONTAL SYSTEMS DURING THE DIAMET CAMPAIGN

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Observations of the Origin and Distribution of Ice in Cold, Warm and Occluded Frontal Systems during the DIAMET campaign

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Abstract

Three case studies in frontal clouds from the Diabatic Influence on Mesoscale Structures in Extratropical Storms (DIAMET) project are described to understand the microphysical development of the mixed phase regions of these clouds. The cases are a kata type cold front, a wintertime warm front and a summer time occluded frontal system. The clouds were observed by radar, satellite and in-situ microphysics measurements from the FAAM research aircraft. The kata cold front cloud was shallow with a cloud top temperature of ~ -13°C. Cloud top heterogeneous ice nucleation was found to be consistent with predictions by a primary ice nucleation scheme. The other case studies had high cloud tops (below ~40°C) and despite no direct cloud top measurements in these regions, homogeneous ice nucleation would be expected. The maximum ice crystal concentrations and ice water contents in all clouds were observed at temperatures around -5°C. Graupel was not observed, hence secondary ice was produced by riming on snow falling through regions of supercooled liquid water. Within these regions substantial concentrations (10 to 150 L⁻¹) of supercooled drizzle were observed. The freezing of these drops increases the riming rate due to the increase in rimer surface area. Increasing rime accretion has been shown to lead to higher ice splinter production rates. Despite differences in the cloud structure, the maximum ice crystal number concentration in all three clouds was ~ 100 L⁻¹. Ice water contents were similar in the warm and occluded frontal cases, where median values in both cases reached ~ 0.2-0.3 g m⁻³, but lower in the cold front case.

1. Introduction

In the mid-latitudes extratropical cyclones can dominate the synoptic situation and therefore the conditions experienced at the surface. Occasionally these systems bring conditions that cause substantial damage to property and significant loss of life, primarily
through strong winds and excessive rainfall (Leckebusch, et al. 2007; Ulbrich, et al. 2001; Ulbrich, et al. 2003a; Ulbrich, et al. 2003b). Observations of the large scale characteristics of these storms has led to a number of models describing their formation and progression, most notably the Norwegian (Bjerknes 1919; Bjerknes and Solberg 1922) and Shapiro Keyser models (Shapiro and Keyser 1990). The dominant features of these mid-latitude storms are the contrasting air masses forming the frontal boundaries that can bring abrupt changes in weather conditions.

The Norwegian model fails to represent the many variations within a cyclones structure on a smaller scale (Browning 1990). Matejka et al. (1980) reported numerous bands of precipitation associated with the frontal systems of mid-latitude cyclones as part of the CYCLOnic Extratopical Storms (CYCLES) project. A distinctive example of this variability between frontal systems is the existence of Narrow Cold Frontal Rainbands (NCFR) and Wide Cold Frontal Rainbands (WCFR) (Matejka et al. 1980; Houze and Hobbs 1982 and Crosier et al. 2013). Often the terms 'ana' and 'kata' (Bergeron 1937) are used to describe different types of cold frontal systems. The ana type cold front has a rearward component to the Warm conveyor Belt (WCB) flow relative to the surface front. This produces rearward-sloping ascent and the generation of a broad area of precipitation known as the WCFR. However, ascent of warm air forced by the surface cold front can lead to more vigorous convection over a narrow area up to an altitude of around 3km (Browning and Reynolds 1994). Kata cold fronts are the result of dry air intrusions, characterised by a region of air descending from the tropopause region that can overrun the surface front (Browning 1997). This prevents rearward ascent of warm air behind the cold front as the dry air intrusion causes a descending motion in all but the lowest layers of the frontal system (Sansom 1950). It also leads to forward motion of the air above the warm conveyor belt relative to the movement of the cold front. This produces forward sloping ascent and causes the main band of precipitation to become situated ahead of the surface cold front (Browning 1986).

Matejka at al. (1980) investigated the microphysics of different frontal bands and, in some cases, highlighted the importance of microphysical and mesoscale convective processes that had a direct influence on the formation and spatial distribution of precipitation. For example, below the narrow convective updraft of an NCFR, precipitation is suppressed through suspension or upward movement of hydrometeors. Crosier et al. (2013) found production of small secondary ice crystals through rime-splintering in an NCFR accounted for a significant fraction of the ice water content (IWC) and suggested this is important in
altering the precipitation budget and the distribution of latent heat release. The reader is referred to Dearden et al. (accepted) for a consideration of diabatic heating and cooling rates derived from some of the microphysics data that will be presented here. In warm frontal systems Hobbs and Locatelli (1978) examined precipitation cores within the frontal rainbands and found they were linked to ice crystals produced at higher altitudes in 'generating cells' that seeded the stratiform cloud below.

The Diabatic Influence on Mesoscale Structures in Extratropical Storms (DIAMET) project (Vaughan et al. submitted), part of the Storm Risk Mitigation (SRM) aims to advance our understanding of the mesoscale features within mid-latitude storms in order to improve accuracy in predicting them. The overarching theme of the DIAMET campaign is to consider the role of latent heating in the generation of mesoscale potential vorticity (PV) and moisture anomalies in cyclonic storms and the impact these may have on the weather. The aims of the campaign most relevant to the work presented here are the measurement of microphysical properties and variability in mesoscale structures. Here we present data from three intensive observation periods (IOPs) of a cold front, warm front and occluded frontal system during the DIAMET campaign. We discuss the large scale features associated with these systems and analyse the dominant microphysical processes that were observed in each case.

2. Methodology

During Autumn-Winter 2011 and Spring-Summer 2012 a total of 14 Intensive Observation Periods (IOPs) involving ground based and in-situ measurements were carried out as part of the DIAMET project. In all cases in-situ measurements were provided by the UK Facility for Airborne Atmospheric Measurements (FAAM) BAe-146 aircraft. The strategy used to explore the microphysics of each frontal system involved flying a combination of constant altitude runs with profiled ascents and descents that aimed to capture the vertical structure of each system. Particular attention was paid to the temperature zone between -3°C and -8°C where secondary ice production (SIP) takes place (Hallett and Mossop 1974; Crosier et al. 2011; Crawford et al. 2011).

Instrumentation used to measure cloud microphysics on-board the BAe-146 included the Cloud Imaging Probe Models 15 and 100 (CIP-15 and CIP-100, Droplet Measurement Technologies Boulder, USA) (Baumgardner et al. 2001), the Cloud Droplet Probe (CDP-100 Version 2, also DMT) (Lance et al. 2010) and the Two Dimensional-Stereoscopic (2D-
S, Stratton Park Engineering Company Inc. Boulder, USA,) Probe (Lawson et al. 2006). The CIP-15 and CIP-100 optical array probes consist of 64 element photodiode arrays with element resolutions of 15 and 100 µm respectively. We analysed data from measurements of particle size over the range 15-930 µm (CIP-15) and 100-6200 µm (CIP-100). The 2D-S also uses an optical array but has 128 elements each of 10 µm resolution, capable of imaging particles between 10-1280 µm. Particle size histograms were provided at 1 Hz for analysis.

It has been recognised for some time that ice breakup on the inlets of probes creates errors in size distribution and number concentration data that need to be corrected (Field et al. 2006). The magnitude of particle enhancement attributed to shattering is dependent on the probe type and the microphysical properties of the cloud being measured. Jensen et al. (2009) found 2D-S concentrations were enhanced by about an order of magnitude when natural concentrations of small cloud ice were low, but negligible when these concentrations were high. During this campaign the CIP-15 and CIP-100 optical array probes were fitted with Korolev designed tips (Korolev et al. 2001) to minimise shattering of ice particles on the probe housing, which can potentially cause artefacts in the measured size distributions. In addition, software processing removed particle shattering events through the use of Inter-Arrival Time (IAT) filtering (Crosier et al. 2013; Field et al. 2006). It should be borne in mind that not all shattering artefacts may be accounted for with IAT filtering.

Size distribution measurements from the CIP-15 and CIP-100 were merged, providing continuous size distribution data sets covering the size range ~ 15-6200 µm. Where there was significant disagreement between the two probes in their respective size overlap regions, preference was given to the CIP-15 dataset, due to its greater resolution (15 µm) compared to the CIP-100 (100 µm). The CIP-15 and CIP-100 datasets were filtered using an IAT threshold of 1x10^-4 s. Tests to examine the sensitivity of ice particle concentrations to different IAT thresholds showed some evidence of shattering artefacts remaining in the size distributions when the IAT threshold was relaxed and lowered to 1x10^-5 s or IAT filtering was turned off completely. As a result a threshold value of 1x10^-4 s was maintained and used in this work.

The 2D-S probe has a significantly higher resolution and frame rate than the CIP-15 and CIP-100 probes enabling it to provide more detailed images of hydrometeors. The higher resolution (at 10µm) and faster response (> 10 times faster) makes the 2D-S generally more suitable for use in the classification of ice particle habits and in discriminating these.
from water drops in each of the cases described here, particularly as a function of altitude and for rapidly changing regions of glaciation. Identification of liquid droplets and ice crystals is based on analysis of a particles shape and is described in Crosier et al (2011). The ability to discriminate between particle phase allowed calculation of the ice water content using the Brown and Francis (1995) mass dimensional relationship. Secondary ice production through the Hallett-Mossop (H-M) process is a known powerful mechanism of glaciation that occurs when cloud temperatures are between -3°C and -9°C, liquid water is present and existing ice particles come into contact with the liquid droplets to produce riming. In these conditions we frequently observe large increases in ice crystal number concentrations. In order to accurately capture the high concentrations of small ice crystals naturally present in the (H-M) temperature zone, an IAT threshold of $1 \times 10^{-5}$ s (as suggested by Crosier et al., 2013) was used. Lowering the threshold to this value for all 2D-S data, increases the possibility that shattered particles could have contaminated the dataset and artificially increased number concentrations, particularly in regions where larger more delicate ice crystals were seen to have developed. This possibility was investigated by examining IAT frequency plots and testing for changes in ice number concentrations when varying the IAT threshold to $1 \times 10^{-4}$ s and also by turning the filtering technique off, allowing all particles to be accepted. IAT frequency plots suggested a limited contribution from a shattering mode and that a threshold of $1 \times 10^{-5}$ s was appropriate.

Cloud particles in this study were selected for analysis using the 'centre in' method (Heymsfield and Parrish 1978), in which the probe's sample volume does not decrease with increasing particle size as is the case with the 'entire in' acceptance technique. This allows greater numbers of larger particles to be measured within a given sample, improving counting statistics. The size of the particles ($D_p$) was calculated by taking the average of the maximum size in the along ($D_y$) and across ($D_x$) directions. The sizing method is less sensitive to elongated particles than using just $D_x$ and allows more accurate representation of particle size distributions in regions where elongated particles such as columns are the dominant growth habit (Crosier et al. 2013). In addition to the cloud microphysics probes used, the aerosol size distribution was measured using a Passive Cavity Aerosol Spectrometer Probe 100-X (PCASP) (Rosenberg et al. 2012) that measures particles over the range 0.1 µm to 0.3 µm diameter. A limited amount of information from this instrument was used to challenge a primary IN parameterisation (DeMott 2010) for the cold front case.
Ground based remote sensing measurements for the cold front IOP, were provided by the Chilbolton Advanced Meteorological Radar, (CAMRa) (Hogan et al. 2003) at the Chilbolton Facility for Atmospheric and Radio Research (CFARR) in Hampshire, Southern England (51.14° N, 1.44° W). This is a fully steerable 3 GHz dual polarisation Doppler radar. During this flight the BAe-146 aircraft flew along a radial of 240° while CAMRa performed Range Height Indicator (RHI) scans along the same radial. Other data collected at CFARR to monitor the passage of the cold front included atmospheric pressure, temperature, wind speed, wind direction and rainfall rate as measured by a rapid response drop counting raingauge (Norbury and White 1971).

3. Frontal Cases

The microphysics and larger scale characteristics of the three cases will now be described in detail. These were chosen to represent 3 distinct frontal types observed during DIAMET. The cold front case provided an excellent opportunity to use in-situ and ground based remote sensing techniques to observe a frontal system in a state of transition. Warm frontal systems are characterised by areas of broad, predominantly light precipitation. The one presented here provided a case in which detailed microphysics measurements of the vertical structure of the system were made. The selection of a summer-time occluded front, which has a much higher freezing level, allowed us to compare with the microphysics of the other two winter-time cases

3.1 Cold Frontal Case

On 29 November 2011, Met Office surface pressure charts showed a large depression north east of Iceland with a central pressure of 961 mb (Fig. 1). A secondary low pressure system developed, pushing a cold front from west to east across the UK, which eventually reached mainland Europe by 00 UTC on 30 November. This cold front was particularly active over Northern England with a squall line and damage caused by a tornado reported in the Greater Manchester area.

The aircraft departed Exeter Airport (50.44°N, 3.24°W) at ~ 14 UTC and arrived overhead Chilbolton at ~ 1430 UTC. A number of constant altitude and stepped profiled runs were repeatedly performed on a radial of ~ 240° from the CFARR location. The aircraft made measurements for ~ 90 minutes before landing back at Exeter airport around 16 UTC.
Examination of the Met Office rainfall radar composite (Fig. 2) shows that the frontal system exhibited both a WCFR and NCFR earlier in the day, which was eventually dominated by the NCFR with no broad area of precipitation. The NCFR exhibited precipitation rates up to 32 mm hr$^{-1}$. Analysis of the in-situ microphysics measurements, meteorological data, remote sensing and satellite products all support the assumption of a transitioning frontal system. Meteosat Second Generation (MSG) cloud top height and temperature products showed that at 10 UTC cloud top temperatures were close to -30°C over a wide area (Fig. 3a). As the front moved over land, cloud top temperatures increased significantly to around -15°C (Fig. 3b). The increase in cloud top temperatures was associated with the lowering of cloud top heights through suppression of convection by the descending dry air over-running the cold front. The 12 UTC radiosonde from Larkhill, just ahead of the frontal rainband, revealed this dry layer between ~ 850 - 600 mb (Fig. 4c). In addition to analysis of radiosonde data, the aircraft deployed 13 dropsondes off the coastline of south west England. Dropsonde data from ~ 09 UTC revealed a saturated profile to 600 mb before the dew point fell to -60°C at 550 mb (Fig. 4a). As the front approached and moved overland dropsonde data from ~ 1240 UTC revealed the appearance of a dry layer between 800 and 650 mb with dew points in this region falling to -30°C. (Fig. 4b).

The passage of the front over CFARR was well represented in the meteorological data recorded at the site. Surface pressure fell to 994 mb at ~ 15 UTC before recovering following the passage of the cold front while winds veered westerly and temperature fell ~ 5°C by 18 UTC as the cold air mass advanced eastwards. The precipitation rate at CFARR, measured by the droplet counting raingauge, reported values typically below 5 mm hr$^{-1}$, and in this case the precipitation arrived before the surface front, which is consistent with the characteristics of a kata type cold front. The front appeared to be in the later stages of transition from ana to kata type as there was some evidence of slantwise ascent behind the front from the radar data shown in figure 5.

The RHI scans from CAMRa showed a broad convective region (Fig. 5a) between ~ 60 and 80 km advancing towards Chilbolton that eventually split into two separate convective elements (Fig. 5d) with the second of the two regions, around 50 km from CFARR, becoming the most active. This development is supported by the rainfall radar sequence that showed a broader region of precipitation (Fig. 2a,d) that split into two bands (Fig. 2b,e) as the front approached CFARR. The dominant convective feature is represented in the CAMRa data by a narrow band of high radar reflectivity up to 50 dBZ (Fig. 5d) ~ 50
km from CFARR. This feature coincided with the passage of the surface cold front and had an associated signature in the unfolded radial velocity field (Fig. 5f) that represents scattering hydrometeors being transported vertically by the convection driven by the advancing cold airmass. Above the frontal uplift, a region of high differential reflectivity, ~ 4 dB, developed between ~ 60 and 90 km from Chilbolton (Fig. 5b) and grew as the front advanced eastwards (Fig. 5e).

The aircraft made several penetrations through this frontal system, as summarised in Table 1. During segment A1 the aircraft made 2 profiled descents and one straight and level run. The altitude of the aircraft varied from ~ 5.2km to 3.7km and temperatures increased from -16°C to -9°C. During this period the aircraft made some brief penetrations of cloud tops in the frontal system. The flight path during these cloud top penetrations intersected regions where CAMRa detected high differential reflectivity. The in-situ microphysics measurements revealed mixed phase, glaciated and supercooled conditions with swift transitions between them (Fig. 6). The 2D-S reported ice crystal number concentrations of upto ~ 1 L$^{-1}$ likely achieved through a combination of heterogeneous ice nucleation in the relatively high cloud top temperatures of around -13°C and the effects of H-M Secondary Ice Production occurring below. This cloud top temperature is only ~ 5°C lower than that in the Hallett-Mossop (H-M) temperature zone that lies between -3°C and -8°C (Hallett and Mossop, 1974). Radar imagery from CAMRa revealed regions of high reflectivity representing convective elements that provided a lifting mechanism that had been generated by the advancing cold front (Fig 5a). 2D-S particle imagery showed that at cloud top, columnar ice crystals that had presumably been lifted from a H-M generating region below, where high concentrations were being produced through SIP. Interestingly, the images also revealed the presence of heterogeneously nucleated ice crystals, in the form of pristine plates (Fig. 7a). The existence of columnar ice crystals within cloud tops might be expected to lead to an enhancement in the ice crystal number concentration there above that expected from primary heterogeneous ice nucleation alone. This is because first ice within the cloud tops is initiated through primary ice nucleation but subsequently these concentrations may be enhanced as ice crystals produced by SIP are lifted from a H-M zone below into the cloud tops. However in this case data measured by the PCASP was used to calculate the expected Ice Nuclei (IN) concentration according to the parameterisation of DeMott et al. (2010) and found the predicted ice crystal concentrations to be about a factor of 2 lower than mean concentrations measured in a cloud top region during a ~ 15 second period by the 2D-S (~ 0.32 L$^{-1}$). The PCASP iced up during the
flight, limiting the data we could use. The mean aerosol concentration > 0.5 µm was calculated from a 15 second time period below cloud base (~ 9.3 cm⁻³).

Cloud Droplet Probe (CDP) derived Liquid Water Contents (LWCs) of 0.1-0.15 g m⁻³ were present within liquid regions at cloud top. During flight segment A2 the aircraft performed a profiled descent from ~ 3.6 km - 1.8 km where temperatures increased from -7°C to 1°C, intersecting the H-M temperature zone where the imaging probes observed large needle and columnar ice crystals in much higher concentrations than those observed at cloud top (~ 80 L⁻¹) (Fig. 7b). During a straight and level run, within flight segment A3, at 1.8 km, outbound from Chilbolton, temperatures fell from ~ +1°C to -2°C and the aircraft passed through the dominant convective feature, where LWCs reached 0.8 g m⁻³, the highest recorded value at any time during the mission. Imagery from the 2D-S revealed small liquid droplets in this region (Fig. 7d) and the CDP reported a mode droplet diameter of ~ 25µm in concentrations ~ 100 cm⁻³. Information about droplet concentration and size as a function of altitude can be found in figure 22. In this case concentrations were generally < 100 cm⁻³ with median diameters between ~ 15 and 30 µm. Analysis of the LWC profile as a function of altitude (Fig. 8a) showed that significant (> 0.01 g m⁻³) LWCs reached right up to cloud top at ~ 4 km. It may be significant that close to the region where the highest concentration of columns were observed, larger supercooled drops (greater than 80 µm diameter) were present in concentrations of up to ~ 150 L⁻¹, the concentrations of drizzle fell to ~ 1 to 30 L⁻¹ in regions where columnar crystals were in concentrations of ~ 80 L⁻¹ (Fig 9a). Crawford et al. (2011) showed the importance of warm rain processes leading to the formation of droplets of diameter > 80 µm in generating high concentrations of ice crystals. Fig. 9a has been modified to exclude ice number concentrations in the time period between ~ 15 UTC and ~ 1511 UTC. Examination of 2D-S probe imagery revealed misclassification of liquid droplets as ice particles during this period. After this point ice crystals were observed in the probe imagery and this data has been included. It is possible that the freezing of the droplets in this case played an important role in the generation of the high ice concentrations in this region.

Matejka (1980) described the microphysics of convective updraught regions within an NCFR as consisting of small growing cloud droplets that were initiated through vertical ascent forced by the surface front. Significant liquid water (> 0.01 g m⁻³) was associated with this region along with low ice particle number concentrations when compared to areas immediately adjacent to the convective element. The cold front in this study displayed similar characteristics, with a high LWC (~ 0.8 g m⁻³) and, for the most part, low ice
number concentrations. In this particular instance the highest concentrations of ice crystals were observed ahead of the convective feature (Fig. 10) and were likely to have been produced through rime-splintering (Hallett and Mossop, 1974). The convective element created by the movement of the cold front from west to east generated supercooled liquid water up to the cloud tops of this frontal system. In these regions small, pristine ice crystals increased in size through riming and vapour growth, which then began to descend and aggregate. The in-situ image data clearly showed ice particles in this region were aggregating and transitioning to snow particles while CAMRa recorded high differential reflectivity values during RHI scans in this same region. As the crystals continue to fall the ZDR signal decreases due to the particles acquiring more complex shapes through changes in habit and the process of aggregation (Fig 5e). Ice number concentration as a function of altitude from the 2D-S show a steady order of magnitude increase in median ice crystal number concentrations from ~ 1 L$^{-1}$ to 10 L$^{-1}$ as the aircraft descended from cloud top into the H-M temperature zone (Fig. 11a). Here, the snow particles came into contact with supercooled liquid water droplets, leading to SIP through rime-splintering. Size distributions from the merged CIP-15 and CIP-100 dataset (Fig. 12a) reveal the high concentrations of small ice crystals ~ 100 µm present during segment A2 while flying through an active area of SIP. This contrasts with lower concentrations of small ice crystals highlighted in a size distribution recorded close to cloud top during segment A1. Note that the ice particle size distributions include all particles classified as ice even in the presence of supercooled drops. Due to the relatively warm cloud tops, this system does not display the typical reduction in ice number concentrations with decreasing altitude, associated with aggregation, that might be expected. The cloud top temperatures of ~ -13°C in this case is close to the main region of secondary ice production between -3°C and -9°C. The ice particles generated in this temperature zone dominate the ice crystal number concentration throughout the depth of the cloud. Percentile plots of ice water content as a function of altitude (Fig. 8b) show that the highest contribution to the ice water content occurred at ~ 3 km, where higher concentrations of ice crystals were observed than in any other region of the cloud system.

Crosier et al. (2013) observed a lack of graupel when investigating an NCFR and suggested that snow particles were the likely sites that produced the riming required for secondary ice production. In this case graupel was also not observed in regions where ice enhancement through secondary ice production was taking place, or in the areas surrounding them. The presence of a component of forward motion relative to the kata cold
front in this case is likely to have directly influenced the spatial distribution of precipitation. Growing ice crystals at cloud top were pushed ahead of the convective feature before descending (Fig. 5e), leading to glaciation of this region through depletion of liquid water droplets via the Wegner-Bergeron-Findeisen process and riming. 2D-S probe imagery reveals snow particles in close proximity to liquid droplets (Fig. 7c). The riming that took place on these snow particles, in the temperature range between -3°C and -9°C, is the likely mechanism by which ice crystal number concentrations were enhanced ahead of the surface cold front. Further details of the cloud ice spectra and their parameterisation in models together with detailed modelling of the diabatic processes in this case may be found in the companion paper Dearden et al. (2014)

3.2 Warm Frontal Case

At 00 UTC on Monday 12 December 2011, Met Office surface pressure analysis charts showed a very unsettled North Atlantic with several low pressure centres (Fig. 1c). By 00 UTC on Tuesday the 13th an intense mid latitude cyclone had developed with a central pressure of 952 mb lying to the west of Scotland (Fig. 1d). Satellite imagery (Fig. 13c-d) revealed a characteristic comma pattern to the cloud structure associated with this mid-latitude cyclone. Eventually the warm front passed across southern England and the system matured, with an occlusion forming over northern parts of the UK. The focus of this study was the warm front, and to investigate this, the BAe-146 operated off the coasts of southwest England and south Wales as the front approached land.

Radar images showed a broad area of rainfall consisting of predominantly light precipitation with intensities typically around 5mm hr⁻¹ (Fig. 14). Meteosat Second Generation (MSG) products showed cloud top heights over an extensive area to be about 10.5 km with temperatures at this altitude around -60°C (Fig. 3c-d). In order to study the evolution within these features, a series of profiled descents and runs at constant altitude were performed from approximately 8.5 km altitude, which are summarised in Table 1. Based on MSG cloud top height products, 8.5 km was ~ 2 km below cloud top. Some of the dropsonde data from the aircraft behind the frontal rain band showed an inversion around 700 hPa, likely representing the warm sector air that had been lifted over the colder air ahead of the warm front (Fig. 15b).

During the first profiled descent (B1) the temperature rose from -43°C, at 8.5km, to -13°C at 4.5 km. The 2D-S measured ice crystal number concentrations at the beginning of the
profile between 25-30 L\(^{-1}\), with small irregular ice crystals with mode diameters between 200-300 µm (Fig. 16a) that very likely formed through homogeneous ice nucleation at temperatures below ~ -37°C. Size distributions for the ice particles only (Fig. 12b) from the merged CIP dataset showed the absence of any larger ice crystals at this altitude. The aircraft then performed a SLR (B2) at 4.7 km where temperatures were between -10°C and -13°C. No liquid water was detected at this altitude where ice crystal number concentrations were between 5 and 10 L\(^{-1}\). There was evidence consistent with heterogeneous ice nucleation at this altitude, in the form of pristine plate-like ice crystals (Fig. 16b). The cloud ice microphysical structure however was dominated by aggregates of ice crystals falling from higher in the cloud. The increase in size, in the absence of liquid water, was mostly due to aggregation as the ice crystals fell, leading to a reduction in number concentration but a general increase in size to a mode diameter of ~ 650 µm, as illustrated in fig. 12b. Reductions in ice crystal number concentrations due to aggregation have been described and quantified in previous studies (Cardwell, Field and Choularton, 2003; Crosier et al 2013) and this is very likely to be the mechanism observed here. The ice particle size distributions recorded during run B2 (Fig. 12b) showed a decrease in smaller ice crystal concentrations compared to the high altitude run (B1) and the presence of higher concentrations of larger aggregated ice crystals ~ 650µm as well as the appearance of ice particles > 1000µm in size.

Percentile plots of ice crystal number concentration (Fig. 11c), mean diameter (Fig. 11d) and ice water content (Fig 8d) as a function of altitude show the change in the microphysics between runs B1 and B2. During the highest altitude run (B1) at a temperature of ~ -43°C ice crystal number concentrations ~ 25 L\(^{-1}\) were observed. With decreasing altitude the process of aggregation gradually increased the mean size while some vapour growth of the ice crystals present increased the IWCs. Around 4 km (-15°C) a more significant increase occurred as the median ice crystal diameter increased to ~ 650 µm and the IWC rose to ~ 0.2 g m\(^{-3}\), probably due to the presence of liquid water at this altitude. Riming also contributes to the increase in size, as the existing growing particles sweep out the supercooled liquid water droplets. Measurements of the droplet concentrations and sizes made by the CDP (Fig. 22) showed low concentrations of droplets in this case, with maximum median values ~ 10 cm\(^{-3}\). Median droplet diameters were < 30 µm.

At 2.8 km, the aircraft penetrated the H-M temperature zone (B3), with temperatures ranging from -4°C to -7°C. The microphysical structure in this region was much more
variable than experienced at higher altitudes (Fig. 11c-d). Changing conditions were observed, including the presence of aggregated ice crystals that had descended from above and the presence of liquid water as detected by the CDP. During this run, high ice crystal number concentrations of a columnar nature up to about 80 L$^{-1}$ were detected (Fig. 16c). As in the cold front case above, significant concentrations of drops in excess of 80 µm diameter were observed by the 2D-S in concentrations of around 15-30 L$^{-1}$ in association with the columnar crystals (Fig. 9b). The enhancement in columnar crystal concentration is likely due to secondary ice production taking place through the H-M process, as ice crystals falling from above sweep out the supercooled liquid water droplets and grow through riming. As in the cold front case, a possible role for the larger drops needs to be considered. Analysis of the LWC profile measured from the CDP (Fig. 8c) showed that the less intense convection associated with these types of fronts generally prevents significant liquid water penetrating to above the ~ -5°C level. However in this case significant LWC was still seen to exist up to around 3 km, which was within the H-M temperature zone. The percentile plots representing ice number concentration (Fig. 11c), mean ice particle diameter (Fig. 11d) and ice water content (Fig. 8d) as a function of altitude reveal that at around 3 km, where temperatures increase to ~ -5°C, higher concentrations of ice particles (~ 80 L$^{-1}$) and higher ice water content values (~ 1.2 g m$^{-3}$) were observed than anywhere else within the warm frontal system.

3.3 Occluded Frontal Case

On 15th August 2012 the aircraft performed a number of in-cloud runs to investigate the microphysical structure of an occluding frontal system over and off the coast of Northern Ireland. The synoptic situation over the UK was notably unsettled for the time of year with a deep low pressure system positioned just south of Ireland at 12 UTC (Fig. 1e). Warm and cold frontal systems pushed north across England and Wales with the occluding sector crossing Northern Ireland (Fig. 1f). Analysis of visible satellite imagery showed a mature cyclone in the process of occluding (Fig. 13e-f). The occluded front was eventually positioned through Northern Ireland, Wales and England while further to the south and west the satellite imagery showed a well-defined trailing cold front.

Dropsonde data from the aircraft (Fig. 17c) show that as the aircraft travelled furthest south, behind the surface front (Fig. 17c), a marked dry sector was observed around 600 hPa and water vapour imagery showed this to be dry air wrapping around the cyclone centre behind the cold front, corresponding to the dark band in fig. 18 lying across Ireland.
and wrapping around the centre of the low pressure system. Radar precipitation rates initially showed a broad frontal system with significant variation in the structure and intensity of precipitation across Northern Ireland (Fig. 19). By 15 UTC an intense squall line with precipitation rates reaching 100 mm hr$^{-1}$ stretched across Northern Ireland, through the Irish Sea and over England. Cloud tops over Northern Ireland were as cold as -52˚C (Fig. 3e-f). The FAAM BAe-146 flew microphysics legs at varying altitudes over Northern Ireland and the sea to the north. During one SLR run at ~ 3 km (C4) the aircraft passed directly through a narrow region of intense precipitation where nearby radar reflectivity indicated rainfall rates of ~ 50 mm hr$^{-1}$. However the higher altitude of the aircraft in relation to the height at which the radar reflectivity actually represented meant that no direct association between the microphysical properties and regions of high precipitation rates could be established.

During profiled climb C1 the aircraft reached an altitude of 7.8 km, the highest altitude at which measurements were made during the flight. Temperatures in this region were around -25˚C, and so a significant distance beneath cloud top. This is confirmed by MSG cloud top height products that suggest cloud tops of around 9km. Here small irregular ice crystals were observed (Fig. 20a) in concentrations between 5 and 10 L$^{-1}$ and with a mode diameter between 200 and 300 µm. There was also evidence of liquid water as the CDP detected a liquid water mass of ~ 0.1 g m$^{-3}$. This was supported by 2D-S imagery that revealed small spherical particles together with ice crystals. In this environment, where ice crystals are observed together with liquid droplets, growth of the ice phase will take place through vapour diffusion. The ice crystals present are also likely to aggregate, which will further increase particle size. The average size distribution from the merged CIP datasets from flight segment C1 (Fig. 12c) shows a noticeable absence of any larger particles over 1000µm at this altitude. During profiled descent (C2) the aircraft descended from 6 to 3 km and temperatures rose from -15˚C to 0˚C. Initially large irregular aggregates of ice crystals were observed together with smaller pristine plates and sectored plates that were likely produced through heterogeneous ice nucleation (Fig. 20b).

Percentile plots of ice concentration (Fig. 11e), mean diameter (Fig. 11f) and ice water content (Fig. 11f) as a function of altitude show that between ~ 8 km and 6.25 km the ice microphysical structure changed little. However at ~ 6 km there was a significant increase in median IWC to ~ 0.2 g m$^{-3}$, and a decrease in median ice number concentrations from about 5 L$^{-1}$ to 2 L$^{-1}$. These measurements highlight a transition from smaller ice crystals, closer to cloud top during run C1, to an increasing number of much larger ice crystals.
observed during C2. The merged CIP size distribution during run C2 also shows the reduction in concentrations of smaller ice crystals with a corresponding increase in the number of larger particles > 1000 µm (Fig. 12c). The number concentrations of the crystals during run C2 vary from just a few per litre to, on occasion, 15 L⁻¹. A number of processes probably contributed to the mix of crystal habits and total ice number concentrations observed at each level during C2 including: (i) Aggregates of ice crystals formed at higher altitudes; (ii) Heterogeneous ice nucleation at this altitude; (iii) columns produced through secondary ice production in the H-M temperature zone below. During the flight, crystals were observed that had changed their growth regimes during their lifetime. Ice crystals were occasionally observed that had initially followed a growth regime in temperatures around -5°C, developing into columns, which were then captured by convective elements within the frontal system, and lifted to an environment where temperatures were significantly lower. At this point the growth regime changed, and the columns developed plate like structures on the ends of the original columnar ice crystals, forming capped columns (as illustrated in Fig. 21).

As the temperature increased during profile C2 the aircraft encountered pristine columnar ice crystals created through SIP in enhanced concentrations of around 40 L⁻¹. This was repeated in a subsequent profiled ascent where ice number concentrations again rose to around 40 L⁻¹ in the H-M temperature zone. These ice crystals were seen together with larger aggregates in the presence of LWCs close to 1 g m⁻³. Analysis of the LWC profile as a function of altitude for the entire science flight (Fig. 8e) showed significant amounts of liquid water penetrating to around 5 km where mean temperatures were ~ -10°C. Analysis of CDP data as a function of altitude (Fig. 22) showed median droplet concentrations up to ~ 100 cm⁻³ and periods in which high concentrations > 500 cm⁻³ were observed. The droplet diameter was generally small with median values < ~ 15 µm. During a subsequent run, (C3), the aircraft maintained a constant altitude within the H-M temperature zone and ice particle number concentrations as high as 100 L⁻¹ were recorded (Fig. 20c). Crystal habits included larger aggregates in the presence of liquid and droplets > 80 µm in diameter (Fig. 9c) in concentrations between ~ 30 L⁻¹, with liquid water contents as high as 1 g m⁻³. Fig. 11 shows percentile plots of ice crystal number concentrations, ice crystal mean size and ice water content from this case. It was observed that during descent into the H-M temperature zone, at ~ 4 km, mean ice crystal number concentrations increased to around 10 L⁻¹, while median ice crystal diameter fell to ~ 300 µm. Periods of secondary ice production increased maximum concentrations in this region to ~ 100 L⁻¹. Percentile plots of the ice water content as a function of altitude (Fig. 8f) show that the highest median ice
water contents (0.25 g m$^{-3}$) occurred at ~ 5.5 km, where temperatures were ~ -10°C. The ice particle size distribution from C3 (Fig. 12c) again showed the highest concentrations of small ice crystals in this region as well as the larger aggregates that were present. All of these observations are consistent with an active H-M zone, enhancing the ice number concentration significantly to above that observed in other regions of this occluded frontal system. Figure 23 shows an example of ice particle size distributions from the 2D-S probe for runs C1 to C3 and confirms the change in the diameter of ice crystals with a reduction in altitude.

4. Discussion

In-situ microphysics measurements showed that in all the cases investigated the production of secondary ice, likely to have been initiated by the H-M process, was evident around -5°C. In these regions the concentration of ice crystals as observed by the 2D-S reached 140 L$^{-1}$. Examination of the LWC profile with altitude reveals that in the cold frontal and occluded frontal cases liquid water was able to penetrate to significantly greater altitudes above the freezing level when compared to the warm front case, with the highest altitude and mass concentrations of liquid water observed in the occluded frontal system. When comparing the summer occluded frontal case to both the winter warm front and the winter cold front cases it was found that liquid water mass was significantly higher in the summer case (Fig. 8e). Examination of the CDP droplet size spectra showed that this increase over the other two cases could be attributed to an increase in the number concentration of small liquid droplets of mode diameter ~ 10 µm. In the winter cases droplet concentrations were lower (generally < 100 cm$^{-3}$) than in the summer case where concentrations reached > 500 cm$^{-3}$. Median droplet diameter in the winter cases reached ~ 25 µm, which was greater than in the summer cases where values were generally < 15 µm (Fig. 22). Despite differences in the distribution of liquid water in each case, significant liquid water was always observed in the H-M temperature zone, where its presence is critical to the powerful secondary glaciation process that takes place through the accretion of supercooled liquid droplets on riming particles. Although the freezing level of the occluded front was ~ 3.5 km (1.5 km higher than the other cases) the convection generated by this system was sufficient to produce significant liquid water to an altitude of ~ 5 km where the temperature of the environment was still within the H-M zone. No evidence for graupel was observed in the H-M temperature zone or adjacent regions on any of the flights. However there were
frequent encounters with rimed aggregates of various ice crystals together with supercooled liquid water in the temperature range -3°C to -8°C.

It may be significant that larger supercooled drops (in excess of 80 um diameter) were found in high concentrations in association with the high concentrations of columnar ice crystals and in the cold front case up to 150 L⁻¹ in a region of supercooled liquid water close to the region of high ice crystal concentrations. Such concentrations of supercooled drops have been reported by Crawford et al (2011) in a rapidly developing convective cloud. The application of a detailed cloud microphysical model in this paper showed that the supercooled drops played a key role in the rapid glaciation of the cloud. The mechanism was that ice particles falling into the Hallett-Mossop zone produced ice splinters that were then captured by the supercooled drops causing them to freeze and become rimers themselves producing further splinters. It was found that this mechanism was able to explain the rapid glaciation of this cloud in the observed timescale.

In the cold front case, relatively warm cloud top temperatures led to initiation of the ice phase through heterogeneous nucleation. In the remaining cases cloud top temperatures were < ~ -37°C. In this environment it is likely homogeneous ice nucleation contributed to the glaciation of the cloud, although heterogeneous ice nucleation via deposition cannot be ruled out. In the cold front case, ice number concentrations of typically up to ~ 1 L⁻¹ within cloud tops were observed, which is in reasonable agreement with the predicted primary IN using the Cooper scheme, as reported in DeMott et al (2010). However, the observed concentrations are two orders of magnitude higher and an order of magnitude lower than predicted by the Fletcher and Meyers schemes respectively (DeMott et al. 2010). Concentrations elsewhere in the cloud reached ~ 80 L⁻¹, which is 1-2 orders of magnitude greater than the predicted primary IN concentration.

In the other two cases, cloud top microphysical regions were well above the H-M temperature zone. There was strong evidence of aggregation processes being important, particularly in the warm front case, resulting in a steady decrease in ice crystal number concentration above the altitude of the -5°C level. For example, observations of ice crystal number concentration (Fig. 11c) and mean size (Fig. 11d) as a function of altitude from the warm front case show a well stratified microphysical system with a continuous decrease in ice crystal number concentration and a corresponding increase in mean particle size (with decreasing altitude) above the H-M temperature zone. Although the summer-time occlusion displayed a similar trend there is much greater variability in these microphysical parameters and this is likely due to strong convection causing intermittent lifting of high
concentrations of these small ice crystals to greater altitudes. In the summer case there was also a substantial depth of cloud below the freezing level and hence warm rain processes will have contributed to the precipitation observed at the surface although no observations were made to quantify this.

In each case, ice water content as a function of altitude (Fig. 8) was calculated using the merged dataset from the CIP-15 and CIP-100. The warm front case initially exhibited little change in ice water content with decreasing altitude, from ~ 8 km to ~ 5 km. Below this level a significant increase in ice water content was observed around 4 km. Percentile plots of liquid water content as a function of altitude show significant liquid water to below 3 km. Ice crystal mass was increasing through the riming process and vapour growth with some of this increased mass lifted to higher altitudes where a notable increase in ice water content was observed just above 4km. In the summer occlusion the significant increase in ice water content occurred much more quickly, taking place at around 6 km. Again this was ~ 1 km above the level where significant liquid water content was observed, suggesting that greater ice water content may have been lifted from regions below.

The depth of cloud in the cold frontal case appears to have significantly affected the ice water content, with maximum median values of ~ 0.025 g m\(^{-3}\) compared to values of 0.25 g m\(^{-3}\) and 0.3 g m\(^{-3}\) in the occluded front and warm front respectively. This may be because ice particles have relatively little time in an environment supersaturated with respect to ice, leading to less growth potential through the B-F process. In all three cases the highest ice water contents were observed where environmental temperatures were ~ -5°C, where secondary H-M ice production through rime-splintering is most efficient.

Radar data from CAMRa (Fig. 5) allowed us to observe the evolution of the cold front as it made progress eastwards. The surface cold front produced a dominant convective feature (~ 50 dBZ) by ~ 15 UTC that delivered significant liquid water to cloud top where the initiation of the ice phase in regions above this feature took place through primary ice nucleation. This was well represented in the ZDR field with high values being returned by pristine plate like crystals. This region of glaciation expanded over time and pushed ahead of the convective feature before the growth of the hydrometeors through riming, aggregation and vapour diffusion took place as they descended into the H-M zone below. By this time the characteristics of the front strongly suggest a kata-type system and the unfolded radial velocity field from CAMRa suggested an element of forward motion in the
cloud tops relative to the lowest layers of the front. This is likely to have pushed the ice crystals initiated here ahead of the surface cold front.

5. Conclusions

With the use of a suite of cloud microphysics instruments, remote sensing measurements and meteorological data we have described and compared the microphysical structures related to the meso-scale characteristics of three contrasting frontal systems.

- Ice formation processes in each case were different. The warm and occluded front presented cloud top temperatures cold enough (< ~ -37°C) for homogeneous and heterogeneous ice nucleation to occur, while the ice phase in the low altitude, relatively warm cloud tops (~ -13°C) of the kata-cold front, was initiated through heterogeneous ice nucleation only.

- The relatively warm cloud top temperatures of the cold front meant that cloud top was in close proximity to the H-M temperature zone below, leading these two areas to become coupled with the ability to directly influence each other. This is highlighted by the presence of columnar ice crystals produced through rime-splintering being present at cloud top, as a result of being lifted up from below. However ice number concentrations at cloud top, measured by the 2D-S, were in reasonable agreement (within a factor of 2) with the predicted primary IN using the DeMott et al. (2010) parameterisation. Concentrations of ice particles elsewhere in the cold front reached ~ 80 L^{-1}, 1-2 orders of magnitude greater than the predicted primary IN concentration at cloud top. This enhancement was due to secondary ice production.

- In all three cases the 2D-S shadow imaging probe measured the greatest ice number concentrations and ice water contents in the H-M temperature zone ~ -5°C. Ice water contents were calculated using the Brown and Francis (1995) mass dimensional relationship. The integrated size distribution from the CDP was used to calculate liquid water contents and concentrations of larger droplets > 80 µm were determined by the 2D-S shadow imaging probe. In all cases significant liquid water
contents (> 0.01 g m$^{-3}$) and concentrations (~ 10 - 150 L$^{-1}$) of larger supercooled drops > 80 µm diameter were observed and this is likely to have facilitated the rime-splintering mechanism and contributed directly to generating these high mass and number concentrations.

- The dynamics of the kata cold front had a direct influence on the distribution of precipitation and location of the most significant area of SIP. This may have occurred as the component of forward motion in these types of fronts carried ice crystals at cloud top ahead of the surface cold front generating the observed patterns. Vertical motions were inferred for the cold front using data from the 3GHz advanced meteorological radar at CFARR. Velocity data from the aircraft was unavailable due to the probe icing up.

- Heavily rimed snow particles were frequently observed and are likely to have provided the sites on which rime-splintering took place.

- The most extensive study to date into splinter production as a function of rimer velocity covered the range 1.5 - 12 m s$^{-1}$ (Saunders and Hosseini, 2001). However the terminal velocity of snowflakes composed of ice crystals exhibiting various growth habits has been shown to be outside of this range (< 1.5 m s$^{-1}$) (Heymsfield 1972; Langleben 1954). In all the cases presented here it is probable that accretion of supercooled water on snow particles travelling at low velocities is the mechanism by which ice crystal number concentrations are enhanced above levels observed anywhere else in each system. Experimental studies of SIP at low riming velocities are therefore needed to quantify this process. The role of supercooled larger drops in the H-M temperature zone also needs further investigation.
Acknowledgements

This work was supported by the Natural Environment Research Council and the Met Office. The core aircraft data was contributed by the Facility for Airborne Atmospheric Measurement.

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CHAPTER FIVE

LATENT HEATING: A CASE STUDY OF A WINTERTIME UK COLD FRONT

The cold frontal system described in the paper titled "Observations of the origin and distribution of ice in cold, warm and occluded frontal systems during the DIAMET campaign" was subject to further analysis relating to diabatic heating and cooling by C. Dearden, P. J. Connolly, G. Lloyd, J. Crosier, K. N. Bower, T. W. Choularton and G. Vaughan (2014). This paper has been accepted for publication in the DIAMET special issue of the Monthly Weather Review. My contribution to this paper involved providing the microphysics datasets used for calculation of latent heating and cooling rates and discussion of the content of the paper throughout. The results of this investigation are described in the following chapter.

5.1 Introduction

The impact of cloud microphysical processes on the characteristics of cloud systems is complex and far-reaching. Latent heating and cooling has the ability to modify the development of mid-latitude cyclones but the complexity of the microphysics processes that influence the magnitude and distribution of latent heat release presents a significant challenge for numerical weather prediction. For example, the growth habit of ice particles in clouds is governed by temperature and supersaturation (Bailey and Hallett, 2009). This impacts on the rate at which vapour diffusion through the Bergeron-Findeisen process leads to ice crystal growth (Fukuta and Takahashi, 1999), and also on the aggregation efficiency of ice particles (Mason et al., 1994). The processes of growth rate, particle shape and aggregation lead to changes in fall speeds (e.g. Locatelli and Hobbs, 1974) that impact on the ventilation of ice particles and this leads to changes in the magnitude and profile of latent heat release as ice particles grow, causing warming of the cloud. Detailed and accurate measurements of in-situ microphysics are, therefore, crucial not only to our
understanding of cloud microphysics but for the development of improved models for use in weather forecasting.

In this paper we calculated diabatic heating and cooling rates from condensation/evaporation, deposition and riming along the flight path of the FAAM BAe-146 aircraft during the science flight investigating the cold frontal rainband. Latent heating rates were determined with the use of a Lagrangian parcel model which was constrained from the in-situ microphysics measurements that were made during the flight. The model uses the 'ode15s' Ordinary Differential Equations (ODE) solver in MATLAB (Shampine and Reichelt, 1997) to determine the evolution of air parcel characteristics through time. Details of the microphysics represented in the bin and bulk scheme can be found in the appendix. Simulations were initialised using information about temperature, pressure, water vapour mixing ratio and vertical velocity and instantaneous latent heating rates calculated for 10 second periods. Details of these equations can be found in the appendix. Model microphysical variables were initialised using particle size distributions (PSDs) measured during the science flight. The evolution of the cloud microphysics and the partitioning of water in the ice, liquid and vapour phases was dependent on the microphysics schemes used in the model, which included explicit (bin-resolved) or parameterized (bulk) representations. When using the bin microphysics scheme the size range and bin width was chosen to represent the microphysics instrument from which the size distributions were measured. For example, the Cloud Droplet Probe (CDP) in the case of measurements of the liquid phase and the CIP-15 or CIP-100 (or a composite of the two) for the ice phase. The bin sizes were 15 µm and 100 µm for the CIP-15 and CIP-100 respectively and varied between 1 and 2 µm for the CDP. When using the bulk scheme gamma and negative exponentials were fitted to the observed PSDs. The impact of the processes such as vapour diffusion and riming were then observed on the evolution of the PSDs.

5.1.2 Calculation of latent heating rates

The change in ice or liquid mass along the aircraft flight track over 10s periods was used to calculate the mass mixing ratio tendency $dq/dt$, in kg kg\(^{-1}\) s\(^{-1}\). From the change in mass we could calculate instantaneous heating rates for the different latent heating processes that include Condensation;
\[
\frac{dT}{dt} \Big|_{\text{con}} = \frac{dq}{dt} \bigg|_{\text{con}} \frac{L_v}{cp_m}
\]

Deposition;
\[
\frac{dT}{dt} \bigg|_{\text{dep}} = \frac{dq}{dt} \bigg|_{\text{dep}} \frac{L_s}{cp_m}
\]

and riming,
\[
\frac{dT}{dt} \bigg|_{\text{rim}} = \frac{dq}{dt} \bigg|_{\text{rim}} \frac{L_f}{cp_m}
\]

where \(L_v\), \(L_s\) and \(L_f\) are latent heat coefficients for vaporization, sublimation and fusion respectively. \(cp_m\) is the specific heat of moist air at constant pressure.

### 5.2 Results

This section describes the results of the latent heating/cooling calculations made using the parcel model. These have been separated into sections focussing on condensation, deposition, sublimation, riming and melting. A summary of some of the profiles and runs that took place during the science flight can be found in Table 2.

<table>
<thead>
<tr>
<th>Log</th>
<th>Time (UTC)</th>
<th>Direction along radial relative to Chillbolton</th>
<th>Temperature</th>
<th>Location of aircraft with respect to frontal system</th>
<th>Summary of observed microphysics</th>
</tr>
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<tr>
<td>Run 10</td>
<td>1414 - 1426</td>
<td>Inbound straight and level from west</td>
<td>Just below freezing level</td>
<td>Through the frontal rainband and into shallower cloud ahead of surface cold front</td>
<td>Liquid with ice falling from above</td>
</tr>
<tr>
<td>Profile 11</td>
<td>1510 - 1515</td>
<td>Outbound descent from cloud top to cloud base</td>
<td>267K - 275K</td>
<td>In the warm sector ahead of surface cold front</td>
<td>Evidence of Hallett-Mossop splinters</td>
</tr>
<tr>
<td>Run 13</td>
<td>1515 - 1521</td>
<td>Outbound straight and level along cloud base</td>
<td>274K - 271K</td>
<td>Flew through convective feature at leading edge of front</td>
<td>Large jump in liquid water content in region of high w</td>
</tr>
<tr>
<td>Run 14</td>
<td>1527 - 1537</td>
<td>Inbound straight and level just below cloud top</td>
<td>265K - 266K</td>
<td>Transitioning from stratiform region to top of convective cells</td>
<td>Brief period of primary ice (large dendrites and aggregates) at 1528 UTC</td>
</tr>
<tr>
<td>Profile 13</td>
<td>1542 - 1545</td>
<td>Outbound descent from cloud top to middle of cloud</td>
<td>266K - 268K</td>
<td>In the warm sector ahead of surface cold front</td>
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</tr>
<tr>
<td>Run 15</td>
<td>1545 - 1557</td>
<td>Outbound straight and level, middle of cloud</td>
<td>268K initially, dropping to 265K</td>
<td>In the cloudy region behind the main convective rainband</td>
<td>Evidence of sublimating ice crystals</td>
</tr>
</tbody>
</table>

Table 2: Description of the runs and profiles used for calculation of heating rates in this paper.
5.2.1 Condensation

Instantaneous latent heating rates attributed to condensational growth of droplets were calculated for Runs 10 and 13 (Table 2). Fig. 23a presents an RHI scan of radar reflectivity from CAMRa, the flight track of the BAe-146 aircraft is overlaid to indicate the aircraft position within the frontal system.

![RHI scan of radar reflectivity](image)

Figure 23: Plots for Run 10. RHI scan of radar reflectivity (dBZ) from CAMRa at 1414 UTC, with black arrow illustrating the aircraft flight track (a); plots of in-situ data along the flight track showing vertical velocity, ambient temperature and CDP droplet number concentration (b-d), and condensation heating rate from the bin parcel model (e). The vertical velocity was produced by subtracting the mean vertical velocity value for the whole run from each 1 Hz measurement to correct for any systematic offset, following advice from FAAM scientists. In plots (b-e), the blue lines represent average values calculated over 1 km intervals from the 1 Hz data, while the shading represents the variability (± 1 standard deviation).

During Run 10 liquid droplets dominated the microphysics. The concentration of droplets measured by the CDP and the latent heating rates attributed to condensation and evaporation are shown in Figures 23d and 23e respectively. The data points in Figures 23a-e are mean values that were calculated every 8 data points, which related to a distance of approximately 1 km based on the aircraft speed of around 100 m s⁻¹. The thick blue line represents the mean and the light blue shading is the standard deviation of each data point.
Heating rates due to the processes of evaporation and condensation were generally positive (i.e. a warming) with maximum values reaching 10 K hr\(^{-1}\). There were some instances where a cooling effect was calculated, for example at around 90 km from CFARR, the instantaneous latent cooling rate due to evaporation was approximately 5 K hr\(^{-1}\).

During Run 13 (an hour later than Run 10) the frontal system had made a transition to a more discrete, intense band of precipitation and the aircraft flew through the narrow updraft associated with the advancing cold front. Radar reflectivity values (Fig. 24a) shows this intense updraft region around 40 km from CFARR. Unfortunately throughout this run the turbulence probe was affected by icing and was not available for significant periods of time. Therefore, only data points where the turbulence probe was operating are provided in Fig. 24c. The condensational heating rates associated with the passage through a small section of the frontal updraft reach nearly 60 K hr\(^{-1}\) (Fig. 24e). It should be noted that the standard deviation is quite large and this is due to the large variability in the updraft velocity. While the turbulence data is limited for this run, the peak updraft velocity are of a similar order to those reported from other vertical velocity measurements in cold fronts for example Crosier et al. (2013).
Figure 24: Plots for Run 13. RHI scan of radar reflectivity (dBZ) from CAMRa at 1514 UTC (a); close-up of the RHI, zoomed in on the area of interest (b); in-situ measurements along the flight track of vertical wind, with additional black line showing 1 Hz data (c); CDP droplet number concentration, CIP-100 number concentration for particles above 400 µm and relative humidity with respect to liquid, all at 1 Hz frequency (d); and the condensation heating rate from the bin parcel model (e), showing the average values of 1 km intervals ± 1 standard deviation.

5.2.2 Deposition and Sublimation

A consideration of the contribution of ice phase processes to latent heating rates is now presented and its importance to diabatic heating processes compared with the findings for liquid drops that were calculated in the previous subsection. Profile 11 (Table 2) contained high concentrations of ice (~ 35 L⁻¹) relative to previous ones investigating the frontal system. Information relating to Profile 11 is shown in Fig. 25
Figure 25: Plots for Profile 11. RHI Scan of radar reflectivity from CAMRa at 1510 UTC focusing on the flight track region (a); in-situ measurements along the flight track of ambient temperature, with relative humidity with respect to ice in grey shading (b); ice crystal number concentration (c); deposition heating rate from the bin and bulk parcel models assuming spherical ice (d-e). In plots d-e, the blue lines account for the effect of ventilation due to sedimentation, whilst the red line assumed the ice crystals have negligible fall speed with respect to the air parcel (i.e. no ventilation).

During the profile the number concentrations of ice particles reach a maximum around 15 km from CFARR and it is here that the highest heating rates are calculated (approximately 3 K hr$^{-1}$). When comparing these with the heating rates associated with condensational growth the contribution from deposition in this particular region is relatively small. As the profile progressed the aircraft descended into a sub-saturated region with respect to ice that produced a cooling rate of around -1 K hr$^{-1}$. Fig. 25d and 25e compare the results from the bin and bulk schemes respectively and despite some small differences the two schemes were generally found to be in good agreement. The addition to latent heating/cooling by the ventilation effect is also represented in Fig. 25d and 25e. The blue trace assumes ventilation effects due to sedimentation, and the red trace shows heating rates calculated assuming ice crystals have a negligible fall speed with respect to the air parcel i.e. no ventilation. The data show that the contribution to diabatic heating by the ventilation effect
is about 50% during periods of peak heating and cooling, as ice particles are subject to
vapour growth and sublimation, but relatively small at other times.

There is a tendency for the bulk scheme to predict greater heating rates than the bin scheme
is explained through examination of the exponential fits used in this approach compared
with the PSDs used in the bin scheme. Fig. 26a represents the mean PSD during Profile 11
and an exponential fit to the data. It shows that the fit under predicts particles > 3 mm and
slightly over predicts concentrations between about 200 µm and 2 mm. Fig. 26c is the first
moment of the size distribution for Profile 11, which is proportional to the growth rate of
ice crystals through vapour diffusion. It shows that the greatest contribution to the change
in mass of the ice phase is dominated by ice crystals < 2 mm. Above this size range the
exponential fit is very close to the observed PSD values, suggesting that the underestimate
of particles in the size ranges > 3 mm (see Fig. 26a) does not impact significantly on the
heating/cooling rates. The findings suggests that it is slight overestimation of particles in
the size range 200 µm to 2 mm that is the main cause of the over estimation of
heating/cooling rates by the bulk scheme.

![Figure 26: Analysis of the time mean ice particle size distribution for Profile 11 (1511 -
1515 UTC). Observations (from the merged CIP data) are shown in blue; negative
exponential fit in red. Comparison of dN/dD (a); comparison of DdN/dD (b); comparison
of \( \int_{D_{\text{min}}}^{\infty} DN(D) dD \) evaluated as a function of \( D_{\text{min}} \) (c). The latter represents the]
contribution of particles with size ≥ \( D_{\text{min}} \) to the overall growth/evaporation rate by vapour diffusion.

Run 15 (Table 2) took place in sub saturated conditions with respect to water and ice in the cloud layer behind the leading edge of the cold front. Measurements of the microphysics suggested that ice crystals that had formed heterogeneously in the cloud layer above were descending into sub saturated air along the flight track. Fig. 27d and 27e show the calculated sublimation cooling rates for this run. Instantaneous cooling rates, assuming ventilation effects, were as high as 2 K hr\(^{-1}\) around 55 km from CFARR. The impact of ventilation on the cooling rates is also greater when compared to similar results from Run 11 and this is attributed to larger ice crystals that have higher fall speeds. Both bulk and bin schemes generally agree, although there is a slight overestimation of cooling rates from the bulk scheme at times. Negative exponential fits (Fig. 28) to the observed PSDs during Run 15 were found to be appropriate across most of the size distribution, with a slight over estimate of concentrations at smaller sizes < 1000 µm.

Figure 27: As Figure 25 but for Run 15. The RHI scan shown in (a) was taken at 1547 UTC.
5.2.3 The Effects of Ice Crystal Habit

Fig. 25 and 27 present data in which ice crystals have been modelled as simple spheres with diameter equal to their maximum dimension measured by the CIP probes. It is clear from probe imagery though that Runs 11 and 15 contained a great variety of crystal shapes including pristine columns, needles and pristine plates. Fig. 29a and 29b show instantaneous heating and cooling rates for Profile 11 and 15 after taking into account ice crystal shape. When ventilation effects are ignored, heating/cooling rates in both cases, taking into account ice crystal shape, are shown to reduce by approximately a factor of 2 compared to the assumption of spherical ice. When the influence of ventilation is included the heating/cooling rates do increase, as was seen in Fig. 25 and 27, however the magnitude of the increase is reduced from about a factor of 3 to a factor of 2. This suggests that the variation in ice crystal habit plays an important role in determining the magnitude of ventilation effects.
Figure 29: Deposition heating and sublimation cooling rates, plotted as timeseries, from the bin parcel model for Profile 11 (a) and Run 15 (b), accounting for the effects of ice crystal shape. As in Figures 25 and 27, the red line represents calculations performed without the effects of ventilation, and the blue line with ventilation.

5.2.4 Riming

Here we calculate latent heating rates due to the riming process during Run 14. Fig. 30a shows the aircraft track and radar reflectivity. The run took place just below cloud top in the presence of liquid water droplets and planar ice crystals that appeared to show evidence of riming. Fig. 30e shows that calculated heating rates associated with riming are about an order of magnitude less than those for deposition heating. This is because the latent heat coefficient of fusion is smaller than the sublimation/deposition coefficient. 2D-S imagery during Profiles 11 and 13 revealed little evidence of rime growth and this is thought to be due to secondary ice splinters quickly removing any available liquid water through the Bergeron-Findeisen process. The importance of riming to start the Hallett-Mossop process is well established, but it appears the rapid removal of available liquid after this process begins limits the impact of riming as a source of diabatic heating. Only results from modelling studies using the bulk scheme are presented here, however we don't anticipate any significant impacts based on discrepancies in fitting gamma functions to observed PSDs.
5.2.5 Melting

Cooling rates due to melting ice crystals were calculated for Run 10 when the aircraft flew a straight and level leg close to the top of the melting layer. Microphysics measurements from the aircraft and remote sensing measurements from CAMRa suggest that melting ice makes a significant contribution to precipitation intensity. During the early part of Run 10 the coldest cloud tops and highest radar reflectivity values were measured and probe imagery consisted of snow particles and aggregates of columnar crystals. These findings suggest that ice crystals formed at cloud top grew and descended into the flight path region below. The high differential reflectivity values in Fig. 31a represent the melting band, with the aircraft track overlaid. As the run continued towards CFARR no ice was detected and
CAMRa scans show, reduced reflectivity values and no evident melting band that would be represented by high differential reflectivity values.

We calculated the mean cooling rates due to the melting process in a given depth $\Delta z$, with PSDs retrieved from the in-situ measurements used to calculate the flux of ice sedimteting through the top of the melting layer. The melting rate ($\text{kg kg}^{-1} \text{s}^{-1}$) in each size bin $i$ is given by:

$$\frac{dq_i}{dT} m t = \frac{q_i V_i}{\Delta z}$$

(12)  

In Equation 12 $q_i$ is the mass mixing ratio and $V_i$ is the fallspeed of the ice particles (Locatelli and Hobbs, 1974). The melting layer depth $\Delta z$ is determined using information about differential reflectivity from an RHI scan, which locates the position of the melting layer due to the 'bright band'. In this particular case we estimated the melting layer to be between 300 and 500 metres. The CIP-100 probe was used to capture ice crystals that were large enough to descend from the cloud layers above under gravitational settling and we assumed a power law relationship between the mass of an ice crystal $m$ and its size $D$, of the form $m = c_s D^{d_s}$. We also only selected ice particles over 400 µm due to the presence of drizzle that was being classified as ice by the processing software. It is likely we rejected some small ice crystals, but we calculate that this does not have a significant impact on the ice mass flux.

Fig. 31c-d show the melting cooling rates calculated during Run 10 for three different mass-size relationships. Fig. 31c uses coefficients $c_s = 0.037$ and $d_s = 1.9$ (for size $D$ in millimeters) (Locatelli and Hobbs, 1974) (LH74). Fig. 31d uses $c_s = 0.069$ and $d_s = 2.0$, which are the values used in the Met Office microphysics scheme (Wilson and Ballard, 1999) (WB99), and Fig. 31e uses values of $c_s = \pi \rho_i / 6$ (where $\rho_i = 100 \text{ kg m}^{-3}$ and $d_s = 3.0$. The final coefficient values represent spherical ice and are referred to as RH84, due to their use in modelling of a cold frontal rainband by Rutledge and Hobbs (1983;1984). Analysis of the results shows that the choice of mass-size relationship impacts significantly on the cooling due to the melting process. Peak cooling rates are between -0.5 and -2 K hr$^{-1}$ depending on the scheme used (Fig. 31c-d).
Figure 31: Additional plots for Run 10. RHI scan of differential reflectivity from CAMRa at 1414 UTC (a): in-situ measurements along the flight track of ice number concentration above 400 µm from the CIP-100 probe (b), and cooling rates associated with the melting of ice, using mass-size relationships as follows: LH74 (c), WB99 (d) and RH84 (e). The RH84 scheme represents assumption of spherical ice particles.

5.3 Discussion

One of the main aims of this study was to address the suitability of the parcel model to accurately represent the microphysics observed in the cold frontal system. It was found that the assumption of spherical ice crystals was too simplistic, as imagery from the 2D-S revealed that ice crystals exhibited many different habits. Assuming that these particles were spherical led to an over estimation of heating rates of up to approximately a factor of 2 for vapour deposition heating, for example. As shown in the melting case the assumption of spherical particles gave an over estimation of particle mass leading to even greater differences in the cooling rates. This indicates that importance needs to be placed on selection of an appropriate mass-size relationship in models. Some problems with fitting negative exponentials to PSDs were also identified, with the fits tending to slightly over estimate small particles and significantly under estimate larger particles. The difference in
the observed PSDs and the fits to the data led to a slight over estimation of heating/cooling rates. We also find that ventilation of the particles is important for latent heating/cooling rates and is also sensitive to ice crystal habit, due to changes in fallspeed.

**5.4 Conclusions**

We have calculated instantaneous heating and cooling rates from a narrow cold frontal rainband during the DIAMET campaign for liquid and ice phase processes. We find that:

- The liquid phase was associated with the largest diabatic heating rates, through the processes of condensation and evaporation.
- Heating/cooling rates from the ice phase processes of deposition and sublimation were at least an order of magnitude less than the peak heating rates associated with condensation. Heating rates due to riming were an order of magnitude smaller again, and therefore in this particular case the riming process is not thought to have made a significant contribution to diabatic heating.
- Precipitating ice was found to produce a cooling effect through sublimation and melting.
- Bulk schemes that assume spherical ice particles were found to be inadequate in calculating heating and cooling rates, especially for vapour diffusion and melting processes. Bulk schemes should, therefore, be able to account for the complexity of ice crystals shapes in an effort to predict diabatic heating and cooling profiles with greater accuracy.
- The impact of ventilation on calculated heating and cooling rates was to increase them by up to about a factor of 2, although when taking into account ice crystal habit the effect was not as large.
- The use of negative exponential fits to PSDs in the bulk scheme was generally a reasonable representation. However there was a tendency for the fits to underpredict larger particles > 3 mm and slightly overestimate concentrations of smaller particles between 200 µm and 2 mm. Leading to an overestimation of heating rates in some regions of the cold frontal system.
CHAPTER SIX

OBSERVATIONS AND COMPARISONS OF CLOUD MICROPHYSICAL PROPERTIES IN SPRING AND SUMMERTIME ARCTIC STRATOCUMULUS DURING THE ACCACIA CAMPAIGN


The following chapter has been submitted to Atmospheric Chemistry and Physics Discussions. G. Lloyd wrote the paper, carried out data analysis and collected some of the data during the campaign period. J.R. Dorsey, K.N Bower, M.W. Gallagher, A. Kirchgassner and T. Lachlan-Cope were involved in data collection. H. Jones and J. Crosier were involved in data analysis and T.W. Choularton supervised the research. All authors commented on the paper.
Observations and Comparisons of Cloud Microphysical Properties in Spring and Summertime Arctic Stratocumulus during the ACCACIA campaign.


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Abstract

Measurements from four case studies in spring and summer-time Arctic stratocumulus clouds during the Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaign are presented. We compare microphysics observations between cases and with previous measurements made in the Arctic and Antarctic. During ACCACIA, stratocumulus clouds were observed to consist of liquid at cloud tops, often at distinct temperature inversions. The cloud top regions precipitated low concentrations of ice into the cloud below. During the spring cases median ice number concentrations (~ 0.5 $\text{L}^{-1}$) were found to be lower by about a factor of 5 than observations from the summer campaign (~ 3 $\text{L}^{-1}$). Cloud layers in the summer spanned a warmer temperature regime than in the spring and enhancement of ice concentrations in these cases was found to be due to secondary ice production through the Hallett-Mossop (H-M) process. Aerosol concentrations during spring ranged from ~ 300-400 $\text{cm}^{-3}$ in one case to lower values of ~ 50-100 $\text{cm}^{-3}$ in the other. The concentration of aerosol with sizes, $D_p > 0.5 \ \mu\text{m}$, was used in a primary ice nucleus (IN) prediction scheme, DeMott et al. (2010). Predicted IN values varied depending on aerosol measurement periods, but were generally greater than maximum observed median values of ice crystal concentrations in the spring cases, and less than the observed ice concentrations in the summer due to the influence of secondary ice production. Comparison with recent cloud observations in the Antarctic summer (Grosvenor et al., 2012), reveals lower ice concentrations in Antarctic clouds in comparable seasons. An enhancement of ice crystal number concentrations (when compared with predicted IN numbers) was also found in Antarctic stratocumulus clouds spanning the Hallett-Mossop (H-M) temperature zone, but concentrations were about an order of magnitude lower than those observed in the Arctic summer cases, but were similar
to the peak values observed in the colder Arctic spring cases, where the H-M mechanism did not operate.

1.0 Introduction

The Arctic is a region that has experienced rapid climate perturbation in recent decades, with warming rates there being almost twice the global average over the past 100 years (ACIA, 2005, IPCC 2007). The most striking consequence of this warming has been the decline in the extent and area of sea ice, especially in the warm season. The lowest sea ice extent and area on record were both observed on 13 September 2012 (Parkinson and Comiso, 2013) and despite some uncertainty, ice-free Arctic summers could become a reality by 2030 (Overland and Wang, 2013). The underlying warming is very likely caused by increasing anthropogenic greenhouse gases and arctic amplification, which is a well-established feature of global climate models (GCMs) (see for example IPCC 5th Assessment Report 2014). However, the details of Arctic climate are complex with interactions between the atmospheric boundary layer, cloud, overlying sea-ice and water leading to a number of feedback mechanisms. These interactions are not well understood due to variability in the spatial and temporal extent of feedback mechanisms, and the fact that those that are included in GCMs may not be accurately parameterised (Callaghan et al., 2011). Clouds play an important role in a number of proposed feedback processes that may be active in the Arctic (Curry et al., 1996; Walsh et al., 2002). Arctic clouds are the dominant factor controlling the surface energy budget, producing a mostly positive forcing throughout the year, apart from a brief cooling period during the middle of summer (Intrieri et al., 2002a). These clouds affect both the long-wave (year-round) and short-wave (summer-only) radiation budgets, and influence turbulent surface exchange. Cloud microphysical influence on cloud radiative properties depends on the amount of condensed water and the size, phase and habit of the cloud particles (Curry et al., 1996). These factors are controlled in part by the Cloud Condensation Nuclei (CCN) and Ice Nuclei (IN) concentrations and properties. Very low aerosol concentrations in the Arctic can result in clouds with properties differing greatly from those at mid-latitudes (Tjernström et al., 2008). A paucity of observations in the Arctic means that neither the aerosol processes, nor cloud properties are well understood or accurately represented within models, with the result that aerosol and cloud-forcing of Arctic climate is poorly constrained.

In the Arctic lower troposphere low cloud dominates the variability in Arctic cloud cover (Curry et al., 1996), with temperature and humidity profiles showing a high frequency of
one or more temperature inversions (Kahl, 1990) below which stratocumulus clouds form. During the Arctic summer, therefore, these low clouds often consist of multiple layers, with a number of theories describing their vertical separation (Herman and Goody, 1976; Tsay and Jayaweera, 1984; McInnes and Curry, 1995a). Such cloud layers have been observed during different seasons but the relationship between temperature and the formation of ice in them is not well understood. Jayaweera and Ohtake (1973) observed very little ice above -20 °C, but Curry et al. (1997) observed ice to be present in clouds at temperatures between -8 < T < -14 °C during the Beaufort Arctic Storms Experiment (BASE). It is possible that the large variation in temperature at which glaciation is observed is caused by changes in the concentration and composition of aerosol (Curry, 1995). Recent work, such as in the Arctic Cloud Experiment (ACE) (Uttal et al., 2002) has improved our knowledge of Arctic mixed-phase clouds, which dominate in the coldest 9 months of the Arctic year. ACE reported that clouds were mainly comprised of liquid tops, tended to be very long lived and continually precipitated ice. The longevity of these clouds might be considered unusual as the formation of ice leads to loss of water through the Bergeron-Findeison process. More recently the Mixed-Phase Arctic Cloud Experiment (M-PACE, 2004) investigated the Arctic autumn transition season. M-PACE was conducted on the North slope of Alaska, in the area to the east of Barrow (Verlinde et al., 2007). Again predominantly mixed-phase clouds were observed with liquid layers present at temperatures as low as -30 °. Remote sensing studies also showed that ice was generally present in low concentrations, mostly associated with precipitation shafts, however, there was also evidence of light snow below thicker layer clouds. IN concentrations were also measured and observed to be low, consistent with liquid water being observed down to very low temperatures. Here we present detailed airborne microphysical and aerosol measurements made in stratocumulus cloud regions in the European Arctic during the recent Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaigns. We present data from two aircraft during early spring, in March and April 2013, and from a single aircraft during the following Arctic summer, in July 2013.

The objectives of this paper are:

1. To report the microphysics and cloud particle properties of Arctic clouds, and the properties, number and size distributions of aerosols in the vicinity of these
2. To identify the origin of the ice phase in these clouds and to compare ice crystal number concentrations with the parameterisation of primary Ice Nucleus (IN) concentrations of DeMott et al. (2010)
3. To compare the cloud physics in spring and summer conditions and to identify any contributions of secondary ice particle production.

4. To compare and contrast the mixed phase cloud microphysics of Arctic clouds with clouds observed in the Antarctic.

2.0 Methodology

The ACCACIA campaigns took place during March-April 2013 and July 2013. They were conducted in the region between Greenland and Norway mainly in the vicinity of Svalbard (and further afield to the south and west of the archipelago). The overarching theme of the project was to reduce the large uncertainty in the effects of aerosols and clouds on the Arctic surface energy balance and climate. Key to the work presented here is an understanding the microphysical properties of Arctic clouds and their dependence on aerosol properties. To this end the FAAM BAe-146 aircraft performed a number flights incorporating profiled ascents, descents and constant altitude runs below, within and above cloud during the spring period. This provided high-resolution measurements of the vertical structure of the cloud microphysics and the aerosol properties in and out of cloud regions. The British Antarctic Survey (BAS) Twin Otter aircraft flew during both campaign periods, providing a subset of the BAe 146 measurements. It was the only aircraft present during the summer period. A total of 9 science flights were conducted during the spring period with complementary flights from the BAS twin otter and 6 flights by the BAS twin otter alone during the summer period.

Two case studies are selected from both the early spring and summer campaigns. The spring campaign case studies were selected for having quite different aerosol loadings within the boundary layer. One was in Arctic air with low total aerosol numbers, while the second had higher aerosol loadings in the boundary layer. Summer flight cases were selected for being the cases with the colder cloud layer temperatures for more direct comparison with the spring campaign cases. Even so, summer case cloud layer temperatures were significantly higher than in the spring cases, and were observed to be in the temperature zone, -3 °C to -9 °C, where a powerful mechanism of secondary ice particle production through rime-splintering, the Hallett-Mossop mechanism, (H-M) (Hallett and Mossop, 1974), is known to operate under particular conditions, and so could greatly enhance ice crystal number concentrations.
2.1 Instrumentation

Instrumentation onboard the Facility for Airborne Atmospheric Measurements (FAAM) British Aerospace-146 (BAe-146, or 146) aircraft used for making measurements of the cloud and aerosol microphysics reported in this paper included: the Cloud Imaging Probe models 15 and 100 (CIP-15 and CIP-100, Droplet Measurement Technologies (DMT), Boulder, USA) (Baumgardner et al., 2001), the Cloud Droplet Probe (CDP-100 Version 2, DMT) (Lance et al., 2010) and the Two Dimensional-Stereoscopic Probe (2D-S, Stratton Park Engineering Company Inc. Boulder, USA) (Lawson et al., 2006). The CIP-15 and CIP-100 are optical array shadow probes consisting of 64 element photodiode arrays providing image resolutions of 15 μm and 100 μm respectively. The 2D-S is a higher resolution optical array shadow probe which consists of a 128 element photodiode array with image resolution of 10 μm. The CDP measures the liquid droplet size distribution over the particle size range 3 < d_p < 50 μm. The intensity of forward scattered laser light in the range 4-12° is collected and particle diameter calculated from this information using Mie scattering solutions (Lance et al., 2010).

A Cloud Aerosol Spectrometer (CAS, DMT) and a Passive Cavity Aerosol Spectrometer Probe (PCASP-100X, DMT) were both used to measure aerosol size distributions onboard the 146. The CAS measures particles in the size range 0.51 < d_p < 50 μm using forward scattered light from single particles in the 4-13° range and backscattered light in the 5-13° range. Particle size can be determined from both the forward and back-scattered light intensity using Mie scattering solutions (Baumgardner et al., 2001). The PCASP is another Optical Particle Counter (OPC) and measures aerosol particles in the size range 0.1 < d_p < 3 μm. In this instrument, particles are sized through measurement of the intensity of laser light scattered within the 35-120° range (Rosenberg et al., 2012). All the above instruments were mounted externally on the FAAM aircraft. Non refractory aerosol composition measurements were provided using an Aerodyne Compact-Time of Flight Aerosol Mass Spectrometer (C-ToF-AMS) whilst aerosol black carbon measurements were provided by a single particle soot photometer (SP-2, DMT). Results from these will be reported elsewhere. Examples of additional core data measurements that were also used in this paper include temperature (Rosemount/Goodrich type 102 temperature sensors) and altitude measured by the GPS-aided Inertial Navigation system (GIN).
Instrumentation on board the Twin Otter Meteorological Airborne Science Instrumentation (MASIN) aircraft, relevant to measurements reported in this paper included: A CDP-100 for drop size distributions; a 2D-S (summer only), both similar to those on the FAAM aircraft; a CIP-25 (as on FAAM except consisting of a 64 element photodiode array providing an image resolution of 25 μm) and core data including temperature measured by Goodrich Rosemount Probes (models; 102E4AL and 102AU1AG for non-deiced, and a de-iced temperatures respectively, similar to those used on the FAAM aircraft) and altitude derived from the aircraft avionics (Litef AHRS) system.

2.2 Data Analysis
During each science flight measurements of aerosol and cloud microphysical properties were made. The techniques used to interpret these data are described below.

Cloud Microphysics Measurements
In the paper, 1Hz data from all cloud and aerosol instruments have been further averaged over 10 second periods for presentation unless peak values, from the 1Hz data are used, as stated. The different flight profiles and straight and level aerosol and cloud sampling runs for all cases are summarised in Table 1. A main focus of this study is the formation of the ice phase in arctic stratocumulus. Measurements from the 2D-S probe have been presented in preference to other 2D probe data due this probes significantly faster response time (by > a factor of 10), and greater resolution. During the spring cases it was possible to combine 2D-S data with measurements from the CIP-100 to extend the cloud particle size range. Analysis of imagery from these Optical Array Probes (OAPs) was used to calculate number concentrations and discriminate particle phase. Identification of irregular particles, assumed to be ice, was achieved through examination of each particles circularity (Crosier et al., 2011). Ice Water Contents (IWCs) were determined using the Brown and Francis (1995) mass dimensional relationship.

All cloud microphysics probes were fitted with “anti-shatter” tips (Korolev et al., 2011) to mitigate particle shattering on the probe tips. However, even with these modifications shattering artifacts may still be present particularly under some cloud conditions and these need to be corrected for (Field et al. 2006). To minimise such artifacts, Inter-Arrival Time (IAT) histograms were analysed in an attempt to identify and remove these additional particles, i.e. by removing particles with very short IATs that are indicative of shattered ice crystals. Crosier et al. (2013) reported that careful analysis of IAT histograms for different
cloud microphysical conditions is needed to determine the most appropriate IAT threshold for best case elimination of such artifacts. For example, in regions of naturally high ice crystal number concentrations, such as in the H-M secondary ice production temperature zone, the minimum IAT threshold may need to be reduced more than is usual so as not to exclude too many naturally generated ice crystals with short IATs. In this study, we found a minimum IAT threshold of 1x10^{-5} s and 2x10^{-5} s for the 2D-S and CIP-15 instruments respectively, to be appropriate IAT values for the majority of cloud region data presented.

It was found that the CIP probes and 2D-S ice crystal number concentrations differed by less than 20% over their common size range. In this paper we present the data from the 2D-S due to its larger size range, higher resolution and faster response time.

2.4. Aerosol Measurements

In each case study, aerosol concentration measurements were used to calculate the predicted primary ice nuclei (IN) concentrations from the DeMott et al. (2010, hereafter D10) parameterisation of primary ice nuclei numbers, which is dependent on the number concentration of aerosol particles with diameters > 0.5 μm. Combined measurements of the aerosol concentration using the PCASP and CAS (for spring), and CAS (for summer), were used from cloud free regions selected by applying maximum Relative Humidity (RH) thresholds. This was done to reduce the contribution of any haze aerosol particles less than 0.5 μm in size growing into the size range at higher humidities and being incorrectly included. The FAAM CAS instrument has a lower size threshold of 0.51 μm. D10 notes that the maximum possible aerosol size that could be measured and included in their D10 parameterization was 1.6 μm. However, due to the size bins utilised by the CAS instrument this upper threshold had to be relaxed to 2 μm, although the extra contribution to the aerosol concentrations used in the calculations is likely to be small. Grosvenor et al. (2012) demonstrated that the scheme is not particularly sensitive to small changes in total aerosol concentrations > 0.5 μm in clean Antarctic regions. Measurements from the higher resolution PCASP were selected from the size range 0.5 μm to 1.6 μm, in keeping with the D10 scheme. The D10 predicted IN concentrations were then compared directly as a function of temperature with the observed ice crystal concentrations. The minimum observed median temperature was input to D10 and predicted IN numbers compared with the maximum observed median ice crystal number concentrations for the clouds during each of the 4 cases. The results are shown in Table 2.
The results of this comparison from all 4 cases can be compared with previous observations of Arctic clouds and with recent aircraft measurements of clouds over the Antarctic Peninsula in the summer (Grosvenor et al., 2012). During the Arctic spring campaign we present one case in which aerosol concentrations were ~ 50-100 cm$^{-3}$ while in the second case presented, concentrations were ~ 300-400 cm$^{-3}$.

3.0 Spring Case 1 - Friday 22 March 2013 (FAAM flight B761)

On this day the FAAM aircraft first flew from Kiruna, Sweden (67.85°N, 20.21°E) to Svalbard, Norway landing at Longyearbyen, (78.22°N, 15.65°E) to refuel. After take-off at ~ 1145 UTC a ~ 2 hour science flight was undertaken to the south east of Svalbard (Fig. 1) before returning to Kiruna. The objective was to investigate stratocumulus cloud in this area, near to the ice edge, and from over ice to open ocean (moving from N to S in the target area). The flight focused on a series of profiled descents and ascents to enable measurements to be made of the cloud layer from below cloud base to above cloud top and into the inversion layer above. During the flight there were 3 significant penetrations through the inversion at cloud top and in each case there was a marked temperature increase of ~5°C. Microphysical time series data for this case are presented, with the relevant runs highlighted in Figure 2. For this case, boundary layer aerosol number concentrations (from the PCASP) were found to be relatively low at ~ 50-100 cm$^{-3}$. Widespread low cloud was observed south and east of Svalbard with winds from the north advecting from over the sea-ice towards open sea. Earlier dropsonde measurements (on the transit into Longyearbyen prior to refuelling) showed surface winds of ~ 3 m s$^{-1}$ increasing to 15 m s$^{-1}$ at 500mb.

3.1 Profiled Ascent A1

During profile A1 the aircraft (travelling south) made a profiled ascent from 300 m above the sea surface, reaching cloud base at 650 m, identified using a Liquid Water Content threshold of LWC > 0.01 g m$^{-3}$, as derived from CDP data. Below cloud base the 2D-S probe revealed low concentrations (< 0.5 L$^{-1}$) of irregular snow (Fig. 3d) particles (mean size ~ 530 μm) that had precipitated from the cloud layer above. As the aircraft climbed through cloud base, temperatures decreased to -11 °C. CDP droplet concentrations ($N_{drop}$) (10 second averaged values) increased to ~ 80 cm$^{-3}$, LWCs peaked at ~ 0.2 g m$^{-3}$ and mean
droplet diameters were ~ 8 μm. Measurements from the 2D-S showed ice crystals with mean size ~ 415 μm in low concentrations, ~ 1 L⁻¹. Images from the 2D-S revealed irregular snow particles with some dendritic habits coexisting with small liquid droplets. As the ascent continued the aircraft encountered a layer containing higher $N_{ice}$ at -14 °C. Ice crystals consisted of snow particles (mean size 350 μm) in concentrations ~ 4 L⁻¹. Probe imagery showed these to be a mixture of large irregular ice crystals, small, more pristine plate-like crystals and some crystals with columnar habits. The highest 10 second mean $N_{ice}$ reached ~ 6 L⁻¹ with peak values ~ 15 L⁻¹. These were observed in a region approximately 500 m below cloud top. Maximum 10 second averaged Ice Water Content (IWC) reached 0.2 g m⁻³ with peaks up to 0.3 g m⁻³ in the same region. Particle images here revealed (Fig 3b) irregular ice crystals together with a few smaller pristine plates. The mid region of this stratocumulus deck also consisted of liquid droplets (mean diameter ~ 13 μm) in concentrations ~ 75 cm⁻³, and LWC ~ 0.3 g m⁻³, with some 1 second integration periods being as high as 0.5 g m⁻³. As the aircraft approached cloud top, where the lowest temperature recorded was -19.5 °C, $N_{ice}$ reduced to ~ 0.5 L⁻¹ with mean sizes of 285 μm, however this region was dominated by liquid droplets (mean diameter 17 μm) with $N_{drop}$ up to 95 cm⁻³, and LWC values peaking at 0.7 g m⁻³. Imagery from the 2D-S revealed many small droplets together with numerous small irregular ice crystals in this cloud top region. After measuring the vertical structure of the cloud layer, which was approximately 1 km in depth, the aircraft penetrated cloud top at 1675 m and passed through an inversion layer where the temperature increased to -13 °C.

3.2 Profiled Descent A2

During profile A2 the aircraft (now travelling north) descended from the inversion layer. Cloud top was encountered at 1650 m ($T = -18.6$ °C). The highest values of $N_{ice}$ were observed in the cloud top region, at ~ 4 L⁻¹ with peaks up to 7 L⁻¹ where IWCs were 0.15 g m⁻³. Particles here consisted of small irregular ice particles (mean size ~ 360 μm) that showed evidence of riming, together with small droplets. CDP LWC increased to 0.3 g m⁻³ with $N_{drop}$ ~ 55 cm⁻³ (mean diameter ~17 μm). At an altitude of around 1400 m asl (~ 250 m below cloud top) $N_{ice}$ decreased to ~ 1 L⁻¹, and mean ice particle size increased to ~395 μm. $N_{drop}$ increased to ~ 70 cm⁻³, while mean size decreased slightly (~16 μm). LWCs generally decreased somewhat to ~ 0.2 g m⁻³. As the aircraft descended to an altitude of ~1150 m, $N_{ice}$ increased by approximately a factor of 2 (to ~ 2 L⁻¹). At around 1315 UTC a number of rapid transitions from liquid to predominantly glaciated conditions were
observed in the mid cloud region at 730 m and $T = -12 \, ^\circ\mathrm{C}$. The initial phase change occurred as LWC decreased from 0.2 to 0.01 g m$^{-3}$ while IWCs increased to a peak value of 0.2 g m$^{-3}$ and peak $N_{\text{drop}}$ fell close to 1 cm$^{-3}$. 2D-S imagery (Fig 3c.) highlights these changes taking place as small droplets are quickly replaced by small irregular ice crystals and eventually larger snow particles (mean diameter ~ 610 $\mu$m) that consisted of heavily rimed ice crystals and aggregates, some of which can be identified as exhibiting a dendritic habit. Observations of dendritic ice are consistent with the ice crystal growth habit expected at this temperature level (-12 °C). Three further swift phase transitions were observed as the aircraft approached cloud base. LWC in the liquid dominated regions was between ~ 0.15 and 0.25 g m$^{-3}$ while $N_{\text{drop}}$ peaked at ~ 130 cm$^{-3}$. During the ice phase sections of the transition cycle, mean particle sizes were ~ 615 $\mu$m and $N_{\text{ice}}$ peaked at up to 5 L$^{-1}$. The contribution of these glaciated cloud regions to the IWC was considerable, with values up to 0.1 g m$^{-3}$ recorded. These transitions ended as the aircraft descended below cloud base ($T = -12 \, ^\circ\mathrm{C}$) at 700 m asl, and precipitating snow was observed (mean size ~ 710 $\mu$m).

### 3.3 Profiled Descent A3

Following another ascent, the aircraft performed a profiled descent (A3) from the inversion layer, $T = -13^\circ\mathrm{C}$, penetrating cloud top at 1,569 m asl where $T = -16 \, ^\circ\mathrm{C}$. As the aircraft descended, LWC increased rapidly to 0.9 g m$^{-3}$ at 30 m below cloud top, the highest LWC recorded at any point during the flight. Mean droplet diameters in this region were ~ 23 $\mu$m in concentrations of ~ 90 cm$^{-3}$. 2D-S images revealed many small liquid droplets with a few small (mean diameter 190 $\mu$m) irregular ice crystals (Fig. 3a) with $N_{\text{ice}}$ ~ 1 L$^{-1}$. The region immediately below this cloud top layer, between 1520 and 1275 m, exhibited a steady decline in LWC while droplet concentrations and $N_{\text{ice}}$ maintained similar values to those observed in the cloud top region. Mean ice crystal diameters increased markedly to 520 $\mu$m before LWCs eventually fell to below the threshold value (0.01 g m$^{-3}$), marking the base of an upper layer of cloud. A subsequent cloud layer, 750 m below, was then encountered. In the clear air region separating these two cloud layers temperatures rose by around 5 °C to -11 °C and large (~ 760 $\mu$m) irregular snow particles, some of which exhibited dendritic growth habits, were observed. Precipitation concentrations were generally < 0.5 L$^{-1}$. Mean IWCs in this precipitation zone were ~ 0.01 g m$^{-3}$. The particles observed falling from the higher cloud layer descended into the cloud layer below at 1,275 m asl. In the top of this lower cloud layer ($T = -11^\circ\mathrm{C}$) LWCs rose to 0.4 g m$^{-3}$ with
N_{drop} (mean diameter 15 μm) increasing to ~ 120 cm\(^{-3}\) while N_{ice} increased to ~ 1 L\(^{-1}\), 2D-S probe imagery in this region revealed the presence of larger snow particles (mean diameters ~ 815 μm). As the aircraft descended further, LWCs gradually decreased while N_{drop} remained fairly constant before reaching cloud base at 280 m, (much closer to sea level than in profiles A1 and A2). Below cloud base precipitating snow (mean particle size ~ 625 μm) was observed.

4.0 Spring Case 2 – Wednesday 3 April 2013 (FAAM flight B768)

The FAAM aircraft departed Longyearbyen at around 11 UTC and conducted measurements to the NW of Svalbard to investigate low-level clouds over sea ice as well as the transition to deeper more convective type cloud as the aircraft moved away from the ice edge and over warmer water (moving from NW to SE in the target area - Fig 1). A low pressure (1004 mb) region was centred south of Svalbard with an associated band of cloud and precipitation. To the NW of Svalbard, within the measurement area, surface winds were E-NE and < 10 m s\(^{-1}\). Measurements revealed an airmass containing significantly more aerosol than in Spring case 1, with PCASP concentrations typically ~ 300-400 cm\(^{-3}\) in the boundary layer. During the flight the aircraft made two distinct saw tooth profiles through the cloud layer and into the inversion above cloud top where temperatures in each instance increased by ~ 2°C. Figure 4 shows time series of the microphysical measurements made during this science flight.

4.1 Profiled Descent B1

Flying NW, the aircraft performed a profiled descent from the inversion layer (T = -16.5 °C) into cloud top, ~ 1550 m asl, where the measured temperature was -17 °C. LWCs rose to a peak value of ~ 0.9 g m\(^{-3}\) and N_{drop} (mean diameter ~ 15 μm) peaked at ~ 320 cm\(^{-3}\). The highest values of N_{ice} never exceeded 0.5 L\(^{-1}\) in this cloud top region and imagery from the 2D-S probe revealed many small droplets with isolated small (mean size ~ 223 μm) irregular ice crystals (Fig 5a). After descending through this brief cloud top region N_{ice} increased to ~ 0.5 L\(^{-1}\). As the aircraft descended over the next 500 m mean droplet concentrations gradually increased from 300 cm\(^{-3}\) to 370 cm\(^{-3}\) with mean diameters decreasing slightly to 12.5 μm. LWCs fell from 0.7 g m\(^{-3}\) to 0.2 g m\(^{-3}\) over the same period and temperatures increased from -17.5 °C to -13.5 °C. N_{ice} values remained fairly constant and IWCs peaked around ~ 0.5 g m\(^{-3}\). 2D-S imagery showed ice crystals (mean diameter
295 μm) to be mainly dendritic in nature. During the last 160 m depth of the cloud before cloud base, $N_{\text{ice}}$ remained similar to the mid-cloud region. However, concentrations of liquid droplets measured by the CDP showed greater variability. Peaks in number concentrations reached as high as $430 \text{ cm}^{-3}$, with rapid changes down to as low as $110 \text{ cm}^{-3}$.

The aircraft passed cloud base at 700 m asl encountering low concentrations ($< 0.5 \text{ L}^{-1}$) of precipitating snow. Interestingly, as the aircraft continued its descent (to 50 m asl) a significant increase in $N_{\text{ice}}$ was observed ($T = -9^\circ\text{C}$), with 10 second mean values of 2 L$^{-1}$ and 1 second peak values of 4 L$^{-1}$. Images from the 2D-S revealed (fig. 5d) snow precipitation co-existing with small columnar ice crystals. CDP LWC was very low, $< 0.01 \text{ g m}^{-3}$, however examination of the 2D-S imagery showed the presence of spherical drizzle droplets, larger than the maximum detectable size of the CDP. Size distribution data from the 2D-S in this region revealed an additional mode dominated by these smaller columnar ice crystals, typically 80 μm in size. As the aircraft ascended again, these higher concentrations of ice crystals diminished before cloud base was reached again at ~ 850 m asl.

4.3 Profiled Ascent B2

During profiled Ascent B2 (prior to profile descent B1 above) the aircraft climbed from below cloud base at 190 m ($T = -5 ^\circ\text{C}$) travelling initially through snow precipitation in concentrations peaking at ~3 L$^{-1}$ (mean diameter 420 μm). Images revealed dendritic ice crystals that had descended from the cloud layer above (fig. 5c). IWCs in this region peaked at 0.025 g m$^{-3}$. Cloud base during this profile was less well defined than in later ascents with variable LWCs and droplet number concentrations before a more defined cloud base was encountered at 1010 m. $N_{\text{drop}}$ then increased rapidly to 270 cm$^{-3}$ (mean diameter ~ 12.5 μm) while LWCs increased more gradually to ~ 0.4 g m$^{-3}$. $N_{\text{ice}}$ through this region showed a decline to < 0.1 L$^{-1}$, and consisted of precipitating snow particles with a mean diameter of 430 μm. Closer to cloud top (1410 m) ice crystal number concentrations increased, to peak values of ~ 1 L$^{-1}$. Images (fig. 5b) showed smaller crystals (mean diameter ~ 370 μm) at this higher altitude, with evidence of hexagonal habits and peak values of IWC ~ 0.04 g m$^{-3}$. Droplet concentrations towards cloud top were similar to lower in the cloud, while LWCs increased to 0.6 g m$^{-3}$ and mean droplet diameter increased to ~ 15 μm. The coldest temperature reached within the cloud layer was -18 °C, but cloud top (at ~ 1530 m) was warmer by 1 °C. A further increase of 1°C was observed as the aircraft ascended through the inversion layer. The depth of this cloud layer (520 m)
was significantly less than that observed during the previous spring case cloud layer penetrations.

4.3 Constant Altitude Runs B3 and B4

During straight and level run (SLR) B3 the aircraft flew below cloud base at 390 m asl to characterise precipitation. During B3 the aircraft briefly traversed a region of low cloud with high $N_{\text{drop}}$ (peaking at ~ 520 cm$^{-3}$) but generally low LWCs (< 0.1 g m$^{-3}$). These cloud droplets were small (mean diameter ~ 6 μm). 2D-S imagery also revealed small drops were present together with snow crystals (mean diameter ~ 370 μm) that were precipitating into these brief regions of low cloud. During B3 temperatures increased from -12 °C to -10 °C. Crystal habits in the out of cloud regions were dominated by aggregates of dendrites and some pristine ice crystals ( ~ 0.5 L$^{-1}$). Here, LWCs were below 0.01 g m$^{-3}$, although the 2D-S also detected drizzle droplets precipitating from the cloud layer above (mean concentration ~ 0.2 L$^{-1}$). Later in B3 the aircraft left its constant altitude and descended to 80 m asl ($T = -8.5$ °C). Mean $N_{\text{ice}}$ increased to ~ 2 L$^{-1}$ with peaks up to 4 L$^{-1}$. There was a corresponding increase in 2D-S droplet concentrations to a mean of ~ 1 L$^{-1}$. 2D-S imagery shows the presence of small columnar shaped ice crystals (similar to those shown in figure 5d), together with larger snow particles and drizzle droplets. CDP LWC was < 0.01 g m$^{-3}$ in this region, since the larger drizzle droplets measured by the 2D-S were outside the CDP size range. In this region of enhanced $N_{\text{ice}}$, just above the sea surface, IWCs, which were generally < 0.01 g m$^{-3}$ in the below cloud base region, increased to peak values of 0.04 g m$^{-3}$.

At the start of run B4, prior to undertaking a mainly straight and level run (SLR) initially to the NW, the aircraft first descended from the inversion layer ($T = -14$ °C) into the cloud top (1050 m asl). LWC initially rose sharply to a peak of 0.5 g m$^{-3}$ before gradually falling away to a mean value ~ 0.3 g m$^{-3}$. Mean droplet concentrations over a ~ 5 minute period were 340 cm$^{-3}$ (mean diameter 11 μm) and the 2D-S imagery revealed the presence of small droplets together with large snow crystals (mean diameter 730 μm) in concentrations < 0.1 L$^{-1}$ and IWCs of 0.03 g m$^{-3}$. At 1240 UTC a generally cloud free region was encountered and sampled for ~ 4 minutes before re-entering cloud again. During this period the aircraft was turned onto a reciprocal heading at the NW limit of its track. Cloud microphysics measurements revealed this cloud top region to be very similar to the first period during B4. Mean values of LWC over ~ 4 minute period were 0.2 g m$^{-3}$, droplet concentrations (mean diameter ~ 9 μm) were ~ 340 cm$^{-3}$. $N_{\text{ice}}$ while generally less than 1 L$^{-1}$ (IWC ~ 0.01 g m$^{-3}$) showed brief increases (during 1 second integration periods) to 2 L$^{-1}$. 


and IWC values peaked at 0.1 g m\(^{-3}\). 2D-S imagery showed the presence of dendritic ice particles (mean diameter 750 μm) together with small spherical particles, likely to be liquid droplets. Temperatures in the cloud top regions remained fairly constant throughout B4 (between -15 °C and -16 °C). The aircraft flew above cloud top for the remainder of the SE-bound leg, and found there to be no ice particles falling into cloud top from above.

**5.0 Summer Case 1 – Tuesday 18\(^{th}\) July 2013 (Flight number M191)**

The MASIN aircraft departed Longyearbyen airport at ~ 07 UTC to conduct a ~ 2hr science flight to the North of Svalbard (Fig. 1). Extensive low cloud was present in the area with light winds < 5 m s\(^{-1}\) from the North. The objectives of the flight were to measure aerosol concentrations and composition in the vicinity of cloud, together with the microphysical properties of the clouds by undertaking a combination of profiles and straight and level runs through stratocumulus cloud layers to capture the microphysical structure. Time series of data collected during this flight are presented in figure 6.

**5.1 Stepped Run C1**

The BAS aircraft performed a stepped profile (flight segments C1.1 - C1.4) from a cloud top altitude of ~ 3000 m down to 2249 m covering the temperature range -7.5 °C to -2 °C. In total 4 SLRs and 4 profiled descents were carried out during this run. During the first penetration of cloud (run C1.1), \(N_{\text{drop}}\) over a 2 minute period was 240 cm\(^{-3}\). LWCs rose to ~ 0.1 g m\(^{-3}\) and the droplet mean diameter was 10.5 μm. \(N_{\text{ice}}\) was generally very low during this period < 0.25 L\(^{-1}\) with some peaks up to 0.5 L\(^{-1}\). During C1.1 the aircraft maintained an altitude of ~ 3000 m for several minutes. The cloud microphysics remained predominantly stable, with low \(N_{\text{ice}}\) (< 0.25 L\(^{-1}\)) and LWCs ~ 0.01 g m\(^{-3}\). The only notable change was a slight increase in the mean diameter of droplets measured by the CDP to 11.5 μm and a reduction in number concentration to 185 cm\(^{-3}\). At ~ 0900 UTC the aircraft descended ~ 100 m to start run C1.2 (\(T= -6^\circ \text{C}\)), and encountered a cloud sector where \(N_{\text{ice}}\) increased to 2 L\(^{-1}\) with peaks to 5 L\(^{-1}\) (and IWC peaks up to 0.03 g m\(^{-3}\) observed here). 2D-S imagery (Fig 7a) revealed irregular ice crystals and the presence of columnar ice both of which appeared to be rimed. Many small single pixel (10 μm) particles were also measured. These likely represent the small droplets detected by the CDP in this region (mean diameter 13.5 μm) in concentrations of 125 cm\(^{-3}\). Later during C1.2, \(N_{\text{ice}}\) fell to values < 0.25 L\(^{-1}\). The aircraft
performed a profiled descent at the start of C1.3, descending 200 m to ~2720 m (T=-4°C). During the descent, LWCs and droplet number concentrations fell to near zero values while $N_{ice}$ increased to peak values of 5 L$^{-1}$ (and IWC peaked at 0.02 g m$^{-3}$). 2D-S images again revealed the presence of small (mean diameter 255 μm) rimed irregular ice crystals and ice crystals of columnar habit. In the temperature range spanned by this cloud, these observations are consistent with the contribution of secondary ice production (SIP) through rime-splintering. During C1.3 further $N_{ice}$ peaks up to 5 L$^{-1}$ consisting of columnar particles and irregular ice crystals were observed (fig 7b). The liquid phase of the cloud in this region was much more variable than nearer to cloud top. Increases in peak LWCs to 0.01 g m$^{-3}$ were seen together with an increase in droplet number concentrations to ~150 cm$^{-3}$ (mean diameter 13.5 μm). These occurred between periods where LWC values were near zero and the cloud was predominantly glaciated.

During C1.4 the aircraft descended 300 m to 2,450 m (T = -3°C). During this run the time between peaks in $N_{drop}$ increased, while the highest $N_{ice}$ measured during this science flight were observed (peaking at $N_{ice}$ = 35 L$^{-1}$). IWCs peaked at 0.2 g m$^{-3}$, which is significantly greater than values observed elsewhere in this cloud system. 2D-S imagery (fig. 7c) reveals that these high ice crystal number concentrations were dominated by columns (mean diameter 260 μm), which at times were seen together with small liquid droplets. These observations are consistent with SIP through the H-M process.

5.2 Profile C2

Later in this flight, the aircraft performed a sawtooth profile, descending from cloud top at ~3300 m down to a minimum altitude of ~2300 m followed by a profiled ascent to complete the sawtooth. During the descent into cloud top (T = -9°C) LWCs rose sharply to peak values of 0.3 g m$^{-3}$ and $N_{drop}$ (mean diameter 19 μm) increased to 155 cm$^{-3}$. $N_{ice}$ in the cloud top regions peaked at 1 L$^{-1}$. With decreasing altitude, LWC declined gradually to values close to 0.01 g m$^{-3}$. As the temperature increased to above -8 °C, ice crystal number concentrations (mean diameter 210 μm) increased to 5 L$^{-1}$, with peaks to ~12 L$^{-1}$. 2D-S imagery revealed the presence of small columnar ice crystals together with small liquid droplets (CDP mean diameter 8.5 μm) and some irregular ice particles. At 2880 m (T = -6.5°C) the cloud dissipated until the next cloud layer was encountered 200 m below (T= -5°C). In this region CDP LWC and $N_{drop}$ were more variable than in the cloud layer above. Generally LWCs were < 0.1 g m$^{-3}$ with peaks in $N_{drop}$ to ~155 cm$^{-3}$ and transitions
between liquid cloud and predominantly glaciated cloud were observed. \( N_{\text{ice}} \) peaked at 25 L\(^{-1}\) and IWCs peaked at 0.15 g m\(^{-3}\). 2D-S imagery showed many columnar ice crystals, typical of the growth regime at this temperature (\( \sim -5 ^\circ\text{C} \)) and consistent with the enhancement of \( N_{\text{ice}} \) through the H-M process. The aircraft reached its minimum altitude (\( T = -3^\circ\text{C} \)) before beginning a profiled ascent to complete the sawtooth. The cloud microphysics of the lower cloud layer were the same as encountered in the descent leg, but with LWCs at times higher (peaks up to 0.2 g m\(^{-3}\)). Transitions between liquid and glaciated phases were observed again, with a notable period of high \( N_{\text{ice}} \) (\( T = -4 ^\circ\text{C} \)), peaking at \( \sim 35 \) L\(^{-1}\) and with IWCs as high as 0.3 g m\(^{-3}\). 2D-S images again revealed many columnar ice crystals (mean diameter 295 \( \mu\text{m} \)), some of which had aggregated, together with irregular ice crystals and liquid droplets. At 2770 m CDP measurements again indicated the presence of a cloud free layer, but over a reduced vertical extent of 100 m, about half the depth observed in the earlier descent. In this region \( N_{\text{ice}} \) reached 8 L\(^{-1}\) in the presence of larger drizzle droplets (fig 7d). Temperatures in the region were around -4 \( ^\circ\text{C} \). Images from the 2D-S showed the presence of small irregular ice crystals with columnar habits. The higher cloud layer cloud base was penetrated at \( \sim 2870 \) m, and \( N_{\text{drop}} \) increased rapidly to 75 cm\(^{-3}\), while LWCs increased gradually to peak values of 0.25 g m\(^{-3}\) at cloud top (\( T = \sim -6 ^\circ\text{C} \)). \( N_{\text{ice}} \) values were lower than those observed lower in the cloud and generally below 5 L\(^{-1}\). Images of the particles showed the presence of small droplets (CDP mean diameter 18 \( \mu\text{m} \)) together with small irregular ice crystals (mean diameter 115 \( \mu\text{m} \)).

6.0 Summer Case 2 – Wednesday 19 July 2013 (M192)

The BAS aircraft departed Longyearbyen at \( \sim 09 \) UTC intending to investigate cloud microphysics and aerosol properties to the north of Svalbard (Fig. 1). On arrival in the observation area the forecasted cloud was not present so the flight was diverted to the south east of Svalbard to meet an approaching cloud system. Surface pressure charts showed a low pressure system over Scandinavia (central pressure 1002 mb), with a warm front south east of Svalbard that was moving north west. Surface winds in this area were \( \sim 13 \) m s\(^{-1}\) from the north east. In-situ cloud microphysics measurements were made for approximately 1.5 hours in total. To meet the objectives of the flight straight and level runs and saw tooth profiles were performed through the cloud layers. Microphysics time series data from the flight are shown in figure 8.

6.1 Profiled descent D1
Well into the flight, the MASIN aircraft performed a profiled descent from cloud top at 3,700 m to 2,400 m over the temperature range -5.2 °C to 3 °C. At cloud top, LWCs rose to a peak of 0.3 g m$^{-3}$, with peak $N_{\text{drop}}$ (mean diameter 12.5 μm) up to 270 cm$^{-3}$. $N_{\text{ice}}$, initially close to zero, rose to peaks of 6 L$^{-1}$ with IWCs up to 0.1 g m$^{-3}$. 2D-S images (fig. 9a) showed columnar ice crystals (mean diameter 350 μm) in this region, together with liquid droplets. At times swift transitions between predominantly liquid and glaciated conditions were observed. At 3,500 m ($T = -3.5$ °C) the CDP stopped measuring significant values of LWC (> 0.01 g m$^{-3}$) and this appeared to mark a gap region in the cloud layer of approximately 100 m in depth. The 2D-S did detect low $N_{\text{ice}}$ in this region. These were generally below < 0.5 L$^{-1}$. When the aircraft descended into the lower cloud layer ($T = -2$ °C) LWCs increased to peak values of 1 g m$^{-3}$, where $N_{\text{drop}}$ (mean diameter 13.5 μm) increased to values as high as 250 cm$^{-3}$. 2D-S imagery revealed few ice crystals in this region but high drizzle drop concentrations.

At 2,800 m ($T = 0$ °C) a further period of drizzle droplets was observed in the 2D-S imagery. These again appeared stretched and made it impossible to separately identify ice in the data set, so there is no reliable ice crystal mass and number concentration data in this region. At this time, CDP LWCs peaked at 0.4 g m$^{-3}$ and droplet concentrations varied from close to zero to up to ~ 350 cm$^{-3}$. The mean diameter of the droplets measured by the CDP was 10 μm. As the aircraft descended towards its minimum descent altitude large variations in LWCs and droplet concentrations continued to be observed with peaks up to 0.2 g m$^{-3}$ and 420 cm$^{-3}$ respectively.

6.2 Profile D2

Earlier in the MASIN flight, during period D2, the aircraft also performed a number of straight and level runs combined with sawtooth profiles to capture the microphysical structure of the cloud layers present. At 3100 m the aircraft flew a straight and level run below cloud base and encountered a region of snow precipitation at temperatures between -2 °C and – 3 °C. $N_{\text{ice}}$ peaked at 5 L$^{-1}$ giving peaks in calculated IWCs of ~ 0.1 g m$^{-3}$. Probe imagery showed ice crystals (mean diameter 410 μm) dominated by irregular particles, with some evidence of plate like and dendritic structures. During a subsequent profiled ascent up to 3400 m (to begin an extended SLR) the aircraft penetrated cloud base at 3300 m ($T = - 4$°C). By the top of the ascent LWCs rose to ~ 0.1 g m$^{-3}$ with $N_{\text{drop}}$ generally
observed to be between 10 and 50 cm\(^{-3}\) (mean diameter 12 \(\mu\)m). \(N_{\text{ice}}\) in this region was between 0 and 1 L\(^{-1}\) with peaks to 3 L\(^{-1}\) and particles consisted of irregular ice particles, columnar ice and small liquid droplets. The mean diameter of the ice particles in this region was 470 \(\mu\)m. Continuing at 3400 m altitude, the aircraft encountered a break in the cloud layer that lasted for around 1 minute (~ 6 km), before a subsequent cloud layer was observed that had similar LWCs to the previous cloud layer (~ 0.1 g m\(^{-3}\)) but with generally lower droplet concentrations (of mean diameter 17.5 \(\mu\)m); with mean \(N_{\text{drop}}\) values of 15-30 cm\(^{-3}\). \(N_{\text{ice}}\) values in this region were lower than before (< 0.5 L\(^{-1}\)). The sampling of this cloudy region was brief before another gap in cloud was observed that lasted ~ 2 minutes. The end of this second clear region was defined by a sudden transition to columnar ice and small irregular particles (mean diameter 410 \(\mu\)m) in concentrations up to a peak of 4 L\(^{-1}\). This region was mostly glaciated with LWC < 0.01 g m\(^{-3}\). During this SLR there were very swift transitions observed between predominantly glaciated regions containing ice crystals (peaking at 4 L\(^{-1}\)) of a columnar nature, and then mainly liquid regions consisting of low concentrations (< 30 cm\(^{-3}\)) of small liquid droplets (mean diameter 14 \(\mu\)m) and LWCs (~ 0.01 g m\(^{-3}\)) (Fig 9c-d). This predominantly glaciated period ended when the aircraft performed a profiled ascent and \(N_{\text{ice}}\) decreased to < 0.5 L\(^{-1}\) while LWCs increased to a peak of 0.3 g m\(^{-3}\) and \(N_{\text{drop}}\) rose to a maximum of ~ 120 cm\(^{-3}\) (mean diameter 14 \(\mu\)m). The aircraft penetrated cloud top at 3,700 m (\(T = -4.5^\circ\text{C}\)).

After climbing above cloud top, the aircraft performed a profiled descent back into the cloud layer to begin another SLR at 3400 m (\(T = -4.5^\circ\text{C}\)). At cloud top LWCs were ~ 0.2 g m\(^{-3}\) \(N_{\text{drop}}\) peaked at 115 cm\(^{-3}\). \(N_{\text{ice}}\) values were greater than in the previous cloud top region. There were two peaks of up to 15 L\(^{-1}\) with particle mean particle diameters of ~ 370 \(\mu\)m. Images show columnar particles, some of which had aggregated, were present together with small liquid droplets (CDP mean diameter 11.5 \(\mu\)m). The second peak contained columnar ice crystals of a similar size (mean diameter 400 \(\mu\)m). The largest spike in ice concentrations occurred in close proximity to the first peak, with values as high as 20 L\(^{-1}\) observed, while IWCs peaked at 0.15 g m\(^{-3}\). Images showed irregular and columnar ice particles (mean diameter 260 \(\mu\)m) present together with small liquid droplets (CDP mean diameter 12 \(\mu\)m) (fig 9b). After these highs in ice number, concentrations declined to ~ 2.5 L\(^{-1}\) before the aircraft made a short profiled ascent and concentrations rose again to peak values of 10 L\(^{-1}\). At 3550 m cloud dissipated and the aircraft descended through a predominantly clear region before reaching another significant cloud layer at 3450 m (\(T = -4^\circ\text{C}\)). CDP \(N_{\text{drop}}\) and LWCs were variable in this region with 10 second mean values
rising to 145 cm$^{-3}$ and 0.1 g m$^{-3}$ respectively. The droplets were small (mean diameter 8 μm) and ice was almost completely absent during this part of the profile. After an SLR at 3,400 m, the aircraft descended as the cloud layer dissipated but encountered another, more significant layer around 3250 m ($T = -2.5 \, ^\circ C$). LWCs increased to peak values of 0.4 g m$^{-3}$ and droplet concentrations (mean diameter 10.5 μm) increased to a peak of 410 cm$^{-3}$. This cloud layer was again predominantly liquid. A spike in 2D-S concentrations was observed which imagery revealed was again due to drizzle droplets. These data were removed from the ice dataset.

7.0 Primary IN Parameterization Comparison

Ice number concentrations as a function of altitude for science flight periods have been presented and here these observations are compared to calculations of the primary IN concentrations predicted using the $D10$ scheme, using aerosol concentrations (diameter > 0.5 μm) that were measured on each flight as input. DeMott et al. (2010) analysed datasets of IN concentrations over a 14-year period from a number of different locations and found that these could be related to temperature and the number of aerosol > 0.5 μm. The parameterisation provided an improved fit to the datasets and predicted 62% of the observations to within a factor of 2. Table 2 shows mean aerosol concentrations for measurement periods during each case, the input temperature to $D10$, the maximum median ice concentration used for comparison and the predicted IN concentration based on both the PCASP and CAS aerosol measurements (where available). During the spring measurement campaign it was possible to compare the CAS and PCASP probe data sets. Despite some variation in concentrations reported between the two instruments, $D10$ predicted IN values were found to be fairly insensitive to these differences. Grosvenor et al. (2012) highlighted that changes of about a factor of 4 produced a very limited change in the IN concentrations predicted by the scheme.

In spring case 1 the maximum median ice value reached 0.61 L$^{-1}$ so predicted IN values were generally higher (between a factor of 2 and 4) than this median ice concentration observation. However peaks in ice concentrations of up to ~ 10 L$^{-1}$, were also observed (Fig. 2) so on these occasions $D10$ significantly under predicts observed ice number concentrations when compared to these peak values. During spring case 2, maximum median ice concentration values were similar to spring case 1. Secondary ice production was observed close to the sea surface in this case so these higher median concentrations
have been disregarded for the purposes of the D10 primary IN comparison. Aerosol measurements from the CAS were lower than from the PCASP but predicted IN values were in good agreement (less than a factor of 2) with the observed maximum median concentration. The peak concentrations observed during the flight were $\sim 5 \text{ L}^{-1}$ (fig. 4) and as in the first spring case D10 under predicted these peak concentrations by about a factor of 10.

During summer case 1 the minimum cloud temperatures were higher (-10 °C) than in the spring cases as were the maximum median ice concentrations (3.35 L$^{-1}$). The origin of these enhanced concentrations is attributed to SIP, making a direct comparison with the D10 primary IN scheme difficult. Predicted IN concentrations from D10 were found to underestimate the maximum median ice concentrations observed in this summer case, but were in agreement with the concentrations observed near cloud top, at temperatures just colder than those of the H-M zone. Observed ice concentrations in summer case 2 were also higher than in the previous spring cases and similar to the first summer case. The second case had higher minimum cloud temperatures than in the first summer case (-4.3 °C). Due to effect of SIP at this temperature, the D10 scheme predicted primary IN concentrations that were quite low when compared to the natural ice concentrations observed in this summer cloud case.

8.0 Discussion

A summary of the microphysics of all 4 case studies is shown in figures 10, 11 and 12. Figure 10 shows the cloud liquid droplet parameters, figure 11 the ice crystal concentration statistics and figure 12 the ice mass and diameter parameters. In each case (a) is spring case 1, (b) spring case 2, (c) summer case 1 and (d) summer case 2. The yellow lines on the ice plots (Fig. 8) show the approximate location of cloud top and cloud base altitudes deduced from liquid water content measurements exceeding 0.01 g m$^{-3}$ from the CDP. It is notable that droplet concentrations (Fig. 10) are much higher in the second spring case than in the first spring case (max median values ~ 60 and ~ 400 cm$^{-3}$ for spring case 1 and 2 respectively) and this is attributed to differences in aerosol concentrations. $N_{\text{drop}}$ are similar in the two summer cases (max median values 100 - 150 cm$^{-3}$) and lie between the two spring cases.
During the spring cases the mixed phase cloud layers were found to be approximately adiabatic and exhibited generally uniform increases in LWC and droplet diameter (Fig. 10) to liquid cloud tops that were observed to precipitate ice. At and above cloud top, well-defined temperature inversions were present. The ice phase is very likely to have been initiated through primary heterogeneous ice nucleation in the temperature range spanned by these clouds (approximately \(-10 \, ^\circ C > T > -20 \, ^\circ C\)). Generally low concentrations of ice crystals were observed (max median value 0.61 L\(^{-1}\)) (Table. 2), but with peaks up to \(~ 5-10 \, L^{-1}\) in both spring cases (Fig. 11). Cloud top regions consisted of small liquid droplets (median diameter \(~ 25 \text{ and } 15 \, \mu m\) for spring cases 1 and 2 respectively) (Fig. 10a-b), together with small irregular ice crystals (Fig 3a and Fig 5a). In both of these cases, ice crystal diameter increased to maximum values of \(~ 530 \, \mu m\) and \(~ 660 \, \mu m\) respectively (Fig. 12a-b). The variability in ice crystal diameter (fig. 12a-b) shows periods where maximum ice crystal diameters increased to \(~ 2 \, mm\). These crystals were often comprised of a mixture of large rimed irregular particles (Fig. 3 and 5) and dendritic snow crystals.

Median IWC values in the spring cases reached \(~ 0.01 \, g \, m^{-3}\) (Fig. 12a-b), with peak values during case 1 up to \(~ 0.3 \, g \, m^{-3}\) compared with \(~ 0.1 \, g \, m^{-3}\) in case 2. The highest Median LWCs (Fig. 10) were observed at cloud top during spring cases, peaking at 0.3 and 0.5 g m\(^{-3}\) during cases 1 and 2 respectively. While these clouds were seen to be fairly uniform, time series data (Fig. 2 and 4) show some of the variability in the microphysics that was observed during the science flight.

During the summer cases, the cloud layers spanned a higher temperature range (\(-10 \, ^\circ C < T < 0 \, ^\circ C\)) and well-defined temperature inversions at cloud top were less evident. There was a much greater tendency towards there being multiple cloud layers that were shallower and less well coupled. During summer case 2 a significant temperature inversion was observed (Fig. 10d) in the cloud base region, which suggested a de-coupling of the boundary layer and the cloud system above. Liquid cloud top regions with few (generally \(< 1 \, L^{-1}\)) ice crystals, formed through heterogeneous ice nucleation at these temperatures, were observed in both cases (Fig. 11c-d). LWCs in summer case 1 were lower than the spring cases (median values \(< \sim 0.1 \, g \, m^{-3}\)) and similar in shape to the uniform profiles seen in the spring cases. The second summer case had higher median LWCs (up to 0.35 g m\(^{-3}\)) and showed much more variability with a number of increases and decreases in median LWC values with altitude (Fig. 10d). Median cloud top ice concentrations in summer case 1 were similar to the spring cases (\(~ 0.2 \, L^{-1}\)) (fig. 11d), however maximum median values lower down in the cloud reached \(~ 3.35 \, L^{-1}\) (Table 2), about a factor of 14 higher than in the spring
cases. Peaks in ice number concentrations around the -5 °C level reached between 30-40 L\(^{-1}\). During the summer, the clouds spanned the temperature range -3 to -8°C, where a well-known mechanism of secondary ice production operates through splintering during riming; the Hallet-Mossopp process (H-M). The observations in this case, of liquid water together with ice particles at temperatures around -5 °C, are consistent with this process being active and enhancing ice number concentrations (Fig 7 and 9). Time series (Fig. 6 and 8) showed more variation than in the spring cases. Distinct liquid cloud tops were still evident, but at lower altitudes significant variations in LWCs, droplet number concentrations and ice number concentrations were seen together with gap regions where little or no cloud was present. On a number of occasions predominantly liquid conditions were swiftly replaced by regions of high concentrations of columnar ice crystals. Some of these transitions took place over ~ 1 second or horizontal distance of the order 60 m. These rapid fluctuations were attributed to the contributions from the H-M process. The process of glaciation through secondary enhancement of ice number concentrations is likely to have caused some of this increased variability in cloud properties too, with liquid droplets quickly being removed through depletion of liquid water by the ice phase. The cloud layers during summer case 2 spanned a higher temperature range than summer case 1. Cloud tops were around -4 °C, and median ice number concentrations reached maximum values of 2.5 L\(^{-1}\), about an order of magnitude higher than in the spring cases. Time series (Fig. 8) and percentile plots (Fig. 11d) showed peaks in ice number concentrations to ~ 25 L\(^{-1}\) and in these regions probe imagery revealed distinctive columnar ice crystals likely to have grown from splinters produced via H-M, into habits typical of growth at these temperatures around -4 °C. In addition, the formation of high ice concentrations may have led to the dissipation of some liquid cloud regions below cloud top due to consumption of the liquid phase by ice crystals growing by vapour diffusion (i.e. ice crystal growth via the Bergeron-Findeisen (B-F) process (Bergeron, 1935). This is consistent with the observed summer clouds being more broken than the clouds observed during spring. However, as discussed in the introduction, it is also recognised that cloud-radiation interactions may lead to the separation of cloud layers during the Arctic summer.

Comparison of the observed \(N_{\text{ice}}\) with the D10 parameterization of primary ice nuclei numbers revealed that during the spring case 1, maximum median \(N_{\text{ice}}\) was lower than the primary IN concentrations predicted by D10, but similar in spring case 2. Peaks in \(N_{\text{ice}}\) were much higher than the D10 IN predictions, by an amount depending on the aerosol measurement period used as input to D10 (Table 2). In the summer cases the enhancement
of $N_{\text{ice}}$ through the H-M process made a realistic comparison difficult. Despite this difficulty, the first summer case had cloud top temperatures that were just outside the H-M temperature zone ($-10 \, ^\circ\text{C}$) and median $N_{\text{ice}}$ in this region was $\sim 0.2 \, \text{L}^{-1}$, which is within a factor of 2 of values predicted by $D10$ (Table 2). At lower altitudes the increase in cloud temperatures allowed rime-splintering to enhance concentrations to above what would be expected via primary heterogeneous ice nucleation. In the second summer case cloud top temperatures were higher ($-4 \, ^\circ\text{C}$), and enhancement of the ice crystal number concentrations through SIP prevented observations of any first ice by primary nucleation being made. Ice crystal number concentrations were thus enhanced to values above what was predicted by $D10$ throughout the depth of the cloud. Whilst primary ice nucleation is identified as the most important ice forming process in the spring clouds, the summer stratocumulus ice concentrations were dominated by secondary ice production via the H-M process as discussed. Due to this SIP enhancement, ice concentrations in summer reached much higher values than those observed anywhere in the spring cases.

The microphysical structure of the spring and summer stratocumulus layers was found to be consistent with previous observations of arctic clouds. We observed generally low droplet number concentrations with increased concentrations during incursions of higher aerosol loadings. This is consistent with observations by Verlinde et al. (2007). During spring cases, LWCs and liquid droplet size increased uniformly to cloud top, however during summer months the vertical structure of cloud layers was more variable (e.g. Hobbs and Rangno, 1998). During spring cases in particular, liquid cloud tops at distinct temperature inversions continually precipitated low concentrations of ice into the cloud below, which has been observed previously in the Arctic. During the Arctic summer, Hobbs and Rangno (1998) observed generally higher ice concentrations with columnar and needle ice crystals in concentrations of ‘tens per litre’ where stratocumulus cloud top temperatures were between $-4 \, ^\circ\text{C}$ and $-9 \, ^\circ\text{C}$. The summer cases we observed contained median values of $N_{\text{ice}}$ that were 4-6 times greater than we observed in the spring cases. In the spring, the cloud layers were colder than the temperature range within which H-M is active, and accordingly contained peak concentrations of ice closer to predictions from $D10$. In the summer cases, the clouds spanned a warmer temperature range between about 0 $^\circ\text{C}$ and $-10 \, ^\circ\text{C}$, leading to low concentrations of primary ice that when conditions became suitable, were then enhanced through rime-splintering. During the spring we also observed cloud that penetrated into the inversion layer, rather than being capped below it. On
average the cloud top was seen to extend ~ 30 m into the inversion layer over which range the mean temperature increase was ~ 1.6°C.

Changes in aerosol concentrations and composition have been suggested as a possible factor in explaining previous observations of the glaciation of arctic clouds at different temperatures (Curry et al., 1996). During spring case 2 higher concentrations of aerosol were observed when compared to spring case 1. Droplet number concentrations were also much higher in spring case 2, generally 300-400 cm$^{-3}$ in comparison to spring case 1 where concentrations were generally ~ 50-100 cm$^{-3}$. Despite this, no significant difference was observed in the ice number concentrations. However, it should be noted that despite the higher total concentrations, the population of aerosol $> 0.5 \, \mu\text{m}$ was not significantly enriched in spring case 2 compared to the spring case 1. $D_{10}$ has a dependency only on this portion of the aerosol size distribution, so may explain the similar primary ice number concentrations for both spring case studies.

Grosvenor et al. (2012) studied stratocumulus clouds in the Antarctic over the Larsen C ice shelf. These observations contained periods where temperatures were comparable to those in the spring cases studied here. The lower layers of Antarctic cloud were also reported to contain higher concentrations of ice produced via the H-M process, similar to the summer cases that we have discussed. A summary of some of the measurements reported from the Antarctic in Grosvenor et al. (2012) can be found in Table 3. Measurements of cloud regions outside the H-M temperature zone revealed very low ice number concentrations, with maximum values about 2 orders of magnitude lower than those observed in the spring cases reported here. Aerosol concentrations from a CAS probe (similar to the one deployed in this study) reported generally lower concentrations of aerosol particles $D_p > 0.5 \, \mu\text{m}$. The $D_{10}$ IN predictions in the Antarctic were reported to compare better with maximum, rather than mean ice values. A similar result was found in this study where predicted primary IN values were greater than observed median values. However, when comparing with peak ice concentration values the scheme significantly under-predicted these. Grosvenor et al. (2012) discussed the possibility that due to the $D_{10}$ parameterisation being based on mean IN concentrations from many samples, the finding that IN predictions compared well with the maximum values rather than mean values may suggest the scheme was over predicting IN concentrations generally in the Antarctic (for these particular cases at least). In the H-M layer in the Antarctic over Larsen C, ice crystal number concentrations were found to be higher than those observed in colder temperature regimes (not spanning the H-M
temperature range), in keeping with the findings from the Arctic presented this paper. However the concentrations produced by the H-M process in the Antarctic were generally only a few per litre, approximately an order of magnitude lower than those observed during the summer cases in the Arctic.

9.0 Conclusions

Detailed microphysics measurements made in Arctic stratocumulus cloud layers during the early spring and summer, have been presented.

- Two spring and two summer cases were presented. The cloud layers during summer cases spanned a warmer temperature range (\(0 \, ^{\circ}C \geq T > -10 \, ^{\circ}C\)) than in spring (generally \(-10 \, ^{\circ}C \geq T > -20 \, ^{\circ}C\)).

- Spring case 2 had significantly higher aerosol concentrations (\(\sim 300-400 \, cm^{-3}\)) compared to the first spring case (\(\sim 50-100 \, cm^{-3}\)). Despite this difference, ice number concentrations were found to be similar in both spring cases, suggesting the source of the increased aerosol concentrations was not providing additional IN that were efficient over the temperature range \(-10 \, ^{\circ}C > T > -20 \, ^{\circ}C\).

- In the spring cases, cloud layers appeared more uniform with steady increases in LWC and cloud droplet size to cloud top, where low concentrations (< 1 L\(^{-1}\)) of ice were frequently observed to precipitate through the depth of the cloud layer. The small irregular particles observed at cloud top grew to a median diameter \(\sim 500 \, \mu m\) in both cases with peaks in diameter \(> 1000 \, \mu m\) as the crystals descended through the cloud. 2D-S imagery revealed the dominant growth habit to be dendritic in nature. The summer cases consisted of multiple cloud layers that were observed to be more variable than in the spring. However, liquid cloud top regions were still evident and ice was again observed to precipitate into the cloud layers below.

- Maximum median ice number concentrations in the summer cases were approximately a factor of 5 (or more) higher than in the spring cases. This enhancement in the ice number concentrations is attributed to the contribution of secondary ice production through the H-M process. As a result of these higher
concentrations, competition for water vapour is likely have limited the maximum size of the ice crystals in the summer (< ~ 1000 \( \mu m \)).

- This finding suggests that low level summer stratocumulus clouds situated in the H-M temperature zone in the Arctic may contain significantly higher ice number concentrations than in spring clouds due to the temperature range of the former spanning the active H-M temperature zone.

- Predicted values from the DeMott et al. (2010) scheme of primary ice nuclei, using aerosol measurements obtained during the science flights as input, tended to overpredict IN concentrations compared to the observed maximum median ice crystal number concentrations during the spring, but under-predict IN when compared to peak ice crystal concentrations. During the summer cases, due to contributions from secondary ice production, the scheme predicted significantly lower values of ice particles than those observed.

- Grosvenor et al. (2012) observed lower concentrations of aerosol > 0.5 \( \mu m \) in the Antarctic when compared to similar measurements made in the Arctic. They found that IN predictions using \( D_{10} \) agreed better with their observed peak ice concentration values rather than their maximum mean values. They measured approximately an order of magnitude lower primary ice concentrations in summer Antarctic clouds than in our spring Arctic cases, but did observe enhancement through SIP in warmer cloud layers where concentrations increased to a few per litre. These were still about an order of magnitude less than the enhanced concentrations observed in the Arctic summer cases presented here, but were similar to the peak values observed in spring cases over the Arctic (where no SIP was observed).
<table>
<thead>
<tr>
<th>Flight</th>
<th>Run Number</th>
<th>Time (UTC)</th>
<th>Altitude (m)</th>
<th>Temperature (°C)</th>
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<tr>
<td>B768</td>
<td>B1</td>
<td>11:45:16 - 11:54:02</td>
<td>1600 - 50</td>
<td>-17 to -9</td>
</tr>
<tr>
<td>B768</td>
<td>B2</td>
<td>11:38:39 - 11:44:59</td>
<td>50 - 1600</td>
<td>-17 to -4</td>
</tr>
<tr>
<td>B768</td>
<td>B3</td>
<td>12:01:30 - 12:19:08</td>
<td>400 - 50</td>
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<tr>
<td>B768</td>
<td>B4</td>
<td>12:32:20 - 12:48:14</td>
<td>1300 - 1050</td>
<td>-16 to -14</td>
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<td>M191</td>
<td>C1.1</td>
<td>08:53:45 - 09:00:00</td>
<td>~ 2950</td>
<td>~ -7</td>
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<td>M191</td>
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<td>09:00:00 - 09:06:50</td>
<td>~ 2900</td>
<td>~ -6</td>
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<td>M191</td>
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<td>09:06:50 - 09:13:35</td>
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<td>M192</td>
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<td>12:58:58 - 13:06:02</td>
<td>3100 - 3750</td>
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<td>12:19:10 - 12:48:16</td>
<td>3100 - 3750</td>
<td>-5 to -1</td>
</tr>
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**Table 1:** Flight numbers, run numbers, and their associated time intervals, altitude and temperature range for the four ACCACIA case studies presented.
Table 2. Measurements of: aerosol concentrations > 0.5 µm from the CAS and PCASP probes, together with predicted primary IN number using the DeMott et al. (2010) (D10) scheme (with either CAS or PCASP aerosol concentration data as input). Observed minimum median cloud temperatures were input to D10, and IN predictions were compared with observed maximum median ice concentrations.

<table>
<thead>
<tr>
<th>Flight</th>
<th>Max Median Ice (L^-3)</th>
<th>Min Median Temp (°C)</th>
<th>Max RH (%)</th>
<th>CAS Aerosol Conc (cm^-3)</th>
<th>PCASP Aerosol Conc (cm^-3)</th>
<th>Predicted CAS IN value (L^-1)</th>
<th>Predicted PCASP IN value (L^-1)</th>
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<tbody>
<tr>
<td>Case 1a</td>
<td>0.61</td>
<td>-18.7</td>
<td>90.3</td>
<td>0.99 ± 0.25</td>
<td>3.13 ± 1.74</td>
<td>1.02 ± 1.14/0.88</td>
<td>1.80 ± 2.25/1.20</td>
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<tr>
<td>Case 1b</td>
<td>0.61</td>
<td>-18.7</td>
<td>22.16</td>
<td>0.14 ± 0.1</td>
<td>4.94 ± 2.22</td>
<td>0.38 ± 0.50/0.21</td>
<td>2.26 ± 2.72/1.68</td>
</tr>
<tr>
<td>Case 1c</td>
<td>0.61</td>
<td>-18.7</td>
<td>85.43</td>
<td>1.48 ± 0.37</td>
<td>4.04 ± 2.25</td>
<td>1.24 ± 1.34/1.08</td>
<td>2.05 ± 2.55/1.37</td>
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<tr>
<td>Case 2a</td>
<td>0.47</td>
<td>-14.1</td>
<td>69.68</td>
<td>1.50 ± 0.30</td>
<td>3.23 ± 1.68</td>
<td>0.47 ± 0.50/0.43</td>
<td>0.63 ± 0.73/0.47</td>
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<tr>
<td>Case 2b</td>
<td>0.47</td>
<td>-14.1</td>
<td>92.60</td>
<td>2.40 ± 0.32</td>
<td>4.96 ± 2.28</td>
<td>0.56 ± 0.58/0.53</td>
<td>0.74 ± 0.84/0.58</td>
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<td>Case 2c</td>
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<td>-14.1</td>
<td>93.86</td>
<td>2.07 ± 6.57</td>
<td>3.07 ± 1.86</td>
<td>0.53 ± 0.90/</td>
<td>0.62 ± 0.73/0.33</td>
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<tr>
<td>Case 3a</td>
<td>3.35</td>
<td>-10</td>
<td>89.37</td>
<td>0.06 ± 0.07</td>
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<td>0.06 ± 0.07/ -</td>
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<td>3.35</td>
<td>-10</td>
<td>59.66</td>
<td>0.15 ± 0.11</td>
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<td>0.08 ± 0.09/0.05</td>
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<td>Case 3c</td>
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<td>-10</td>
<td>89.79</td>
<td>0.33 ± 0.76</td>
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<td>Case 3d</td>
<td>3.35</td>
<td>-10</td>
<td>89.70</td>
<td>0.48 ± 0.21</td>
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<td>Case 4a</td>
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<td>79.70</td>
<td>3.73 ± 1.03</td>
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<td>Case 4b</td>
<td>2.50</td>
<td>-4.3</td>
<td>73.46</td>
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<td>Mean Ice Conc (L⁻¹)</td>
<td>Max ± std. dev. (60 sec) Ice Conc (L⁻¹)</td>
<td>Temp of Max Conc (°C)</td>
<td>Max RH for Aerosol (%)</td>
<td>Observed Aerosol Conc (cm³)</td>
<td>Predicted IN Value (L⁻¹)</td>
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<tr>
<td>99-i4</td>
<td>0.007 ± 0.002</td>
<td>0.017 ± 0.007/0.005</td>
<td>-13.8</td>
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<td>0.33 ± 0.05</td>
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<tr>
<td>99-i5</td>
<td>0.007 ± 0.001</td>
<td>0.020 ± 0.007/0.004</td>
<td>-16.5</td>
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<tr>
<td>104-i3</td>
<td>0.008 ± 0.002</td>
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<td>60</td>
<td>0.15 ± 0.03</td>
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### Hallett Mossop Zone Ice

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<tr>
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<th>Mean Ice Conc (L⁻¹)</th>
<th>Max ± std. dev. (60 sec) Ice Conc (L⁻¹)</th>
<th>Temp of Max Conc (°C)</th>
<th>Max RH for Aerosol (%)</th>
<th>Observed Aerosol Conc (cm³)</th>
<th>Predicted IN Value (L⁻¹)</th>
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<tbody>
<tr>
<td>100-i1</td>
<td>0.52 ± 0.02</td>
<td>1.28 ± 0.06/0.38</td>
<td>-0.7</td>
<td>75</td>
<td>0.42 ± 0.05</td>
<td>1.9×10⁻⁵</td>
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<td>100-i2</td>
<td>1.14 ± 0.02</td>
<td>3.44 ± 0.11/1.01</td>
<td>-2.3</td>
<td>75</td>
<td>0.42 ± 0.05</td>
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<td>100-i3</td>
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<td>0.42 ± 0.05</td>
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<tr>
<td>100-i4</td>
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<tr>
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</tbody>
</table>

**Table 3:** Table reproduced from Grosvenor et al. (2012) reporting observations of ice number concentrations, aerosol concentrations > 0.5µm and primary IN predictions using the *D10* parameterisation.
Figures

Fig 1: Visible satellite imagery for spring case 1 (a), spring case 2 (b), summer case 1 (c) and summer case 2 (d). Flight areas highlighted by purple boxes for each image.
Fig 2: Microphysics time series for spring case 1. Data includes temperature (°C) and altitude (m) (lower panel) together with 1 and 10 second data sets for CDP liquid water content (g m⁻³) (panel 2 from bottom), CDP cloud particle number concentration (cm⁻³) (panel 3), and ice water content (g m⁻³) and ice number concentrations (L⁻¹) (top panel).
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Fig. 7. Images from the 2D-S cloud probe from summer case 1 for: (a) small irregular ice during C1.2; (b) and (c) secondary ice production during C1.3 and C1.4 respectively, and (d) ice together with drizzle during C2.
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Fig. 10: Percentile plots (50th, 25th, 75th percentiles, whiskers to 10 and 90%) as a function of altitude for LWC from CDP (green), and median droplet number concentration (purple), median droplet diameter (grey) and median temperature (red). Data are averaged over 100 m deep layers. Figs. (a - d) are for Spring Case 1, Spring Case 2, Summer Case 1 and Summer Case 2 respectively.
Fig. 11: Box and whisker plots with 50th, 25th, 75th percentiles, whiskers to 10 and 90% and outliers between 95 and 100% as a function of altitude for ice number concentrations (black) and median temperature (red) (Figs. (a-d) and altitude averages as in Fig. 10 above). The box in yellow provides an indication of the full extent of cloud layers investigated. Figs. (a - d) are for Spring Case 1, Spring Case 2, Summer Case 1 and Summer Case 2 respectively.
**Fig. 12:** Box and whisker plots with 50th, 25th, 75th percentiles, whiskers to 10 and 90% and outliers between 95 and 100% as a function of altitude for ice mass (black) and median ice crystal diameter with outliers between 95 and 100% (blue). (Figs. (a-d) and altitude averages as in Fig. 10 above). The box in yellow provides an indication of the full extent of cloud layers investigated. Figs. (a - d) are for Spring Case 1, Spring Case 2, Summer Case 1 and Summer Case 2 respectively.

**Bibliography**


CHAPTER SEVEN

ICE-LIQUID CLOUD TRANSITIONS MEASURED AT HIGH-ALPINE SITE JUNGFRAUJOCH


G. Lloyd carried out data collection, analysis and wrote the paper. M. Flynn, K.N. Bower, R. Farrington, J.Crosier and P.J. Connolly were involved in data collection. J. Cosier helped with the data analysis T Choularton supervised the research. All authors commented on the draft paper
Ice-Liquid Cloud Transitions Measured at High-Alpine Site Jungfraujoch

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Abstract

During January and February 2014 measurements of cloud microphysical properties over a five week period at the high Alpine site Jungfraujoch, Switzerland were carried out as part of the Ice Nucleation Process Investigation and Quantification project (INUPIAQ). Measurements of aerosol properties at a second, lower site, Schilthorn, Switzerland, were used as input for a primary ice nucleation scheme to predict ice nuclei concentrations at Jungfraujoch and to examine any links between potential ABO’s (aerosol particles of biological origin) via their UV-fluorescent signature and ice concentrations. We have identified frequent, rapid transitions in the ice and liquid properties of the clouds at Jungfraujoch site that led to large fluctuations in ice mass fractions over temporal scales of seconds to hours. During the measurement period we observed, with different instruments, high concentrations of ice particles that exceeded 2000 L⁻¹ in temperatures ≈ -15 °C. We were unable to explain these concentrations using the usual primary ice nucleation schemes, which under predicted ice nuclei by several orders of magnitude. Secondary ice production through the Hallet-Mossop process was not likely to be active as the cloud did not lie in the temperature range associated with this. It is shown that other proposed mechanisms of secondary ice particle production cannot explain the highest ice particle concentrations. Very large variations in the air trajectory to the site were observed resulting in large variations in the number and source of the atmospheric particulate including an event with a high loading of Saharan dust aerosol. There was some evidence to suggest that UV-fluorescent aerosol detected by the WIBS instrument (probably of biological origin) may possibly have been responsible for, or contributed in part, to ice nucleation in the temperature range -10C to -12C, consistent with laboratory studies.
1.0 Introduction

During January and February 2014 the Ice Nucleation Process Investigation and Quantification (INUPIAQ) project took place at three high-alpine sites, Jungfraujoch (46.55° N, 7.98° E), Schilthorn (46.56° N, 7.84° E) and Kleine Scheidegg (46.59° N, 7.96° E), Switzerland alongside the Cloud Aerosol Characterisation Experiment (CLACE) 2014. There are comparatively few measurements of the detail of the microphysics at mountain top sites of clouds in supercooled conditions. Choularton et al. (2008) presented data from the CLACE series of experiments that took place at Jungfraujoch (JFJ) in which ice particle number concentrations and habit were measured with a Cloud Particle Imager (CPI, SPEC Inc., USA) probe and water droplets with a Forward Scattering Spectrometer Probe (SPP-100, DMT, USA) and liquid water content with a Particulate Volume Monitor (PVM-100, Gerber Scientific). They found swift transitions between liquid and glaciated cloud that took place on spatial scales of just a few metres. The presence of these fast transitions in other atmospheric settings has been confirmed during airborne measurement campaigns (e.g. Lloyd et al., 2014, submitted ACPD). As well as very rapid phase transitions measured in the clouds at JFJ there were changes that took place over larger temporal scales (several hours) during the campaign period. Glaciated cloud periods at this high alpine site have been found to contain variable ice number concentrations measured by CPI and other probes, often in the range 1-100 L\(^{-1}\) (Choularton et al., 2008) and sometimes exceeding 1000 L\(^{-1}\) Targino et al. (2009). Under conditions where heterogeneous ice nucleation dominates, the concentration and composition of aerosol particles in the atmosphere is likely to significantly influence the number and variability of ice particles. Measurements of interstitial aerosol and residues contained within liquid droplets and ice crystals at JFJ have been found to be enriched in chemical species such as: mineral dust (Kampphus et al., 2010), lead, complex internal mixtures of silicates and metal oxides, secondary aerosol and carbonaceous material (Ebert et al., 2011) compared to the interstitial aerosol. This previous work suggested that particles from industrial activity containing lead were especially important as Ice Nuclei (IN) with contributions from dust and organic material. It was concluded that admixtures of the anthropogenic component may have been responsible for the increased IN efficiency within mixed phase clouds. Sampling of aerosol at JFJ and testing for IN active particles to determine an IN concentration has also shown significant seasonal variability linked to changes in aerosol source. Chou et al. (2011) used a portable ice nucleation chamber (PINC) to make the first continuous measurements of IN over a period of days at JFJ. The system was operated at a temperature of -31 °C and at relative humidities of 127 % with respect to (wrt) ice and 91
% wrt water. They found a mean IN concentration of 8 L⁻¹ during a winter measurement period and 14 L⁻¹ during the summer. They also identified a Saharan Dust Event (SDE) where IN concentrations were seen to increase to several hundred per litre. During the SDE they found the IN to be correlated with larger aerosol particles > 0.5 μm.

1.1 INUPIAQ

One of the key objectives of INUPIAQ is to confirm the previously observed transitions between ice and liquid clouds and to examine the role of changes in aerosol chemical composition. During the campaign period three sites (JFJ, Schilthorn and Kleine Scheidegg) were utilised, with the aim that the lower Schilthorn site act as an upwind out of cloud site to measure aerosol properties in order to examine the influence on prevailing downwind cloud microphysics at the summit of JFJ.

2.0 Site Descriptions

An overview of the locations of each, and a brief description of the instrumentations deployed, at each site is provided in this section. Figure 1 shows the locations of the three sites that were used during the campaign.

2.1 Jungfraujoch

Jungfraujoch is situated high in the Swiss Alps with measurements taken at the Sphinx laboratory at 3580 m asl (Fig. 1). Baltensperger et al. (1998) reported an annual mean cloud frequency at the site of 37%, making it a suitable location for the study of cloud microphysics.

2.1.2 Instrumentation

The JFJ measurement site was used primarily to measure cloud microphysical and residual cloud particle properties. An overview of all instrumentation at this site can be found in Table 1 and some of these instruments are labelled in figure 2. Measurements of cloud liquid water content (LWC) and droplet size distributions were provided by; a Particulate Volume Monitor (PVM, Gerber Scientific, Inc), which measured LWC only (Gerber, 1991), Cloud Droplet Probe (CDP-100, Droplet Measurement Technologies, DMT) (Lance et al., 2010), Cloud Aerosol Spectrometer (CAS, DMT) (Baumgardner et al., 2001) and a Forward Scattering Spectrometer Probe (SPP-100, DMT) (e.g. Dye and Baumgardner, 1984). The most reliable instrument for liquid water content measurements was the CDP as it was least affected by accumulated snow. It generally agreed well with other instruments during periods when they were free of ice / snow. Images of cloud hydrometeors for use in
ice/liquid phase and habit discrimination were provided by the 3 View - Cloud Particle Imager (3V-CPI, Stratton Park Engineering Company (SPEC) Inc, Boulder, Colorado). This instrument is comprised of a Two-Dimensional Stereoscopic (2D-S) (Lawson et al., 2006) and high frame rate Cloud Particle Imager (CPI, SPEC) probes. Wind velocity measurements were provided by a sonic anemometer (Metek) and temperature and humidity provided by a Vaisala probe. Meteorological measurements were also available from the JFJ WMO weather station at the summit site. Measurements of aerosol number concentration and single particle shape and UV-fluorescent properties were made for part of the campaign with a Wideband Integrated Bioaerosol Sensor (WIBS, Version 4, Kaye et al. 2005, Gabey et al. 2010) that was placed at the site between 5 and 25 February 2014.

2.2 Schilthorn

Schilthorn lies 2970 m asl at the summit of the Bernese Alps. The site was selected to act as the upwind measurement location to examine aerosol properties before they arrived at JFJ.

2.2.1 Instrumentation

A summary of the instruments deployed at this site can be found in Table 2. Core instrumentation included an Aerosol Mass Spectrometer (AMS), for chemical aerosol composition measurements. An Optical Particle Counter (GRIMM Technologies Inc., Model 1.109) for measuring aerosol number spectra over the size range $0.25 < D_p < 32 \, \mu m$. Measurements of aerosol number concentration and fluorescent properties were made for part of the campaign with the use of the WIBS 4 instrument that was present at the site between 26 January and 3 February 2014.

2.3 Kleine Scheidegg

The ski resort of Kleine Scheidegg is situated at a lower altitude than that of the Schilthorn and JFJ sites (2061m) and was used for the positioning of a HALO Photonics LIDAR to provide remote sensing measurements of cloud layers and SDE in the area. Figure 1 Shows the location of Kleine Scheidegg, where the LIDAR was located. This paper focuses on the in-situ measurements of the microphysics and aerosol.

3.0 Data Analysis

In this paper the 2D-S probe, part of the 3V-CPI, was used to analyse particle imagery to produce information about concentrations, size and phase. The 2D-S is an optical array
probe (OAP) that has two identical channels consisting of horizontal and vertical laser beams that illuminate 128 photodiode arrays with an effective resolution of 10 µm. Particles passing through either laser shadow the arrays, and images of these particles can are created over the size range $10 < D_p < 1280$ µm. The probe was fitted with an anti-shatter inlet for airborne sampling (Korolev et al., 2001) to minimise any shattering, however analysis of probe imagery and inter-arrival time (IAT) histograms (Crosier et al., 2011) revealed no characteristic shattered particles and this is likely to be due to the velocity at which this probe was aspirated throughout the campaign at ~ 15 m s$^{-1}$.

Discrimination of ice and liquid particles was based on an image circularity criterion (Crosier et al., 2011), and Ice Water Contents (IWCs) were determined with the use of a mass dimensional relationship (Brown and Francis, 1995). Information on liquid droplets was obtained using the CDP, which detects light scattered by droplets and determines a particles optical diameter over the range $2 < D_p < 50$ µm which was confirmed using a Cloud-Aerosol-Spectrometer with Depolarisation (CAS-DPOL) for the same particle size range. Data from the latter will be reported elsewhere. Datasets from these probes were provided at 1Hz for analysis and the data has been further averaged over 10 seconds. Reported concentrations refer to the 10 second averaged dataset, with peak values reported for the shorter 1 second integration periods. Calculation of ice fractions, critical to identifying transitions between glaciated and liquid cloud, was obtained using information about ice and liquid mass (Eq. 1)

$$\text{Ice Fraction} = \frac{M_{ice}}{M_{drop} + M_{ice}}$$

Eq. 1

Where $M_{ice}$ is the ice mass from the 2D-S probe and $M_{drop}$ is the liquid water content from the CDP.

4.0 Synoptic Conditions

January and February 2014 were exceptionally cyclonic and depressions persistently impacted the Atlantic coastal regions of Western Europe. Generally higher pressure over Eastern Europe led to these systems stalling and frequently drawing up winds from a southerly direction across central Europe. Figure 3 shows the mean sea level pressure fields for Europe during January and February 2014.
4.1 Meteorological Conditions

An overview of the conditions experienced at JFJ and Schilthorn during the January and February measurement period are shown in figures 4 (wind rose) and 5 presenting time series data for temperature, pressure, relative humidity and wind velocity.

5.0 Case Studies

Here we examine data during four measurement periods at JFJ, Schilthorn and Kleine Scheidegg from a suite of cloud microphysics instruments to identify ice-liquid transitions taking place at JFJ during January and February 2014. These transitions were seen to vary in magnitude and temporal scale, and we report the number and size of ice and liquid particles together with ice crystal habit where possible. We use measured aerosol size and concentration as input to the primary IN parameterization scheme of DeMott et al. (2010) to compare predicted IN values at JFJ compared to the number of ice particles measured by the various imaging probes. In one of the cases a dust event originating from the Sahara Desert was identified and the possible implications of this event for cloud microphysics at JFJ were investigated.

5.1 11-12 February 2014 (Case 1)

Over the period between 11 and 12 February 2014 low pressure dominated Western Europe, pushing frontal systems across Switzerland. Temperatures at JFJ during this period were between -10 and -20 °C, with the wind direction predominantly from the north with a period of southerly winds between ~ 12 and 18 CET on 11 February. On these days two distinct cloud periods were observed, the first between 00 CET and 09 CET on the 11 Feb and the second between 02 and 10 CET on 12 Feb. Both of these periods were associated with the passage of cold frontal systems with bands of cloud and precipitation. Figures 6 and 7 shows time series data during both transition periods for wind speed and direction, temperature, ice number concentrations ($N_{\text{ice}}$), IWC’s, droplet number concentrations ($N_{\text{drop}}$), LWC’s and ice fraction for both measurement periods. Observations show two transitions from mostly high ice fractions to low ice fractions over each period, both of which are several hours in length. Measurements showed that ambient conditions remained generally stable with temperatures falling approximately 2 C to -16 °C and -18 °C during transition periods one and two respectively. Wind speeds remained between ~ 5-10 m s$^{-1}$ and from a northerly direction throughout.

During the first transition at 00 CET $N_{\text{ice}}$ initially decreased from 500 to 100 L$^{-1}$ before eventually increasing to peak values between 1500 and 2000 L$^{-1}$ with IWCs rising to
3 g m$^{-3}$ at ~ 02 CET leading to ice fractions generally > 0.7. Despite changes in the ice phase, $N_{\text{drop}}$ and LWCs remained stable around 100-200 cm$^{-3}$ and 1 g m$^{-3}$. Size distributions from the 2D-S, measured during the period containing some of the highest $N_{\text{ice}}$ at 0155 CET (fig. 8a), were found to consist of one mode of small particles at 80 µm. After this period of high ice fractions a decrease in $N_{\text{ice}}$ was observed and by 04 CET concentrations had declined to < 300 L$^{-1}$. After 04 CET $N_{\text{drop}}$ increased significantly to values ~ 500 cm$^{-3}$ with 1 second peak values reaching 1000 and 2000 cm$^{-3}$. Ice fractions responded accordingly and fell to generally < 0.2 after 06 CET. 2D-S size distributions at 06 CET, as ice fraction values were falling, showed a decrease in $N_{\text{ice}}$ that was evenly distributed across the size range of the instrument. Size distributions measured by the CDP at 0155 CET (fig. 8b) during the period of high ice fractions showed a broad distribution with significant concentrations of droplets > 20 µm. At 0600 CET, where some of the highest $N_{\text{drop}}$ values were observed, the distribution had changed markedly with a notable reduction in the numbers of droplets > 20 µm and a significant increase in droplets around 10 µm. Images from the CPI and 2D-S during the period of enhanced ice fractions showed many irregular crystals together with pristine plates and small droplets. Observations of plate-like ice crystals are characteristic of a growth habit in the temperature regime ~ -15 °C.

At the beginning of the second transition period the ice fraction was close to 1 at 03 CET and $N_{\text{ice}}$ was ~ 200 L$^{-1}$ with peak concentrations between 500 and 1000 L$^{-1}$ and IWCs 2 g m$^{-3}$. $N_{\text{drop}}$ was observed to be < 100 cm$^{-3}$ with LWCs generally below a significant detectable value (0.01 g m$^{-3}$ as measured by the CDP). 2D-S size distributions at 03 CET showed two modes in the ice crystal size, one around 80 µm, and a second at 300 µm (Figure 8a). CDP size distributions at 03 CET (fig. 8b) demonstrate the low numbers of small liquid droplets observed, leading to low LWCs and high ice fraction during this period.

Shortly after 0400 CET $N_{\text{drop}}$ rose to values around 400 cm$^{-3}$ (LWCs 0.5 g m$^{-3}$) with peaks to ~ 1000 cm$^{-3}$. Size distributions from the CDP at 0420 CET showed a distinct mode of droplets around 5 µm (Figure 8b). The increase in liquid mass took place in the absence of significant change in the ice phase, with only a slow trend to lower $N_{\text{ice}}$. These factors led to a fall in the ice fraction to values < 0.4 for ~ 1 hour from 0400 CET. Due to the fall in ice fraction being attributed to rising LWC, with little change in the IWC, the ice size distribution from 2D-S at 0420 CET is of a similar order of magnitude to the distribution observed during the high ice fraction period at 03 CET (fig. 8a). Following this transition
to lower ice fractions, $N_{\text{drop}}$ and LWCs remained generally stable while $N_{\text{ice}}$ exhibited a number of increases and decreases. Values of $N_{\text{ice}}$ values peaked between $\sim 2000$-3000 L$^{-1}$ (IWCs $\sim 0.2$ g m$^{-3}$) during one such increase at 0500 CET. This interplay between stable $N_{\text{drop}}$ and changing $N_{\text{ice}}$ is represented by increases and decreases in the ice fraction as the variability of $N_{\text{ice}}$ impacts on the IWC. By 08 CET $N_{\text{ice}}$ fell to a few 10s per litre, while $N_{\text{drop}}$ continued to remain stable. This is highlighted by the ice fraction values, which fell close to zero. Temperatures over this transition period fell approximately 2 °C to -18 °C, wind direction remained from a northerly sector and wind speed was stable between 4 and 8 m s$^{-1}$.

5.2 30 Jan – 31 Jan 2014 (Case 2)

At 01 CET On 30 January 2014 low pressure centred in the Mediterranean sea (central pressure 999 mb) advanced eastwards towards northern Italy, introducing a southerly flow across the Swiss Alps and pushing a warm front into Northern Italy. Between 01 CET 31 January and 01 CET 01 February the warm front stalled, and cloud associated with this system moved into Switzerland as the low pressure centre filled (central pressure increased to 1005 mb) and moved south-eastwards. Precipitation produced by this depression remained confined to southern Switzerland. Over the measurement period two ice-liquid transitions were observed. Time series data for this measurement period (parameters plotted as in case 1) can be found in figures 9 and 10.

During the first transition period (between 06 CET and 00 CET 31 Jan) the temperature rose from -19 °C to -15 °C and wind speed, from a southerly direction, increased from 10 m s$^{-1}$ to a maximum of 20 m s$^{-1}$ at 17 CET. The start of period one consisted of both low IWCs ($< 0.01$ g m$^{-3}$) and $N_{\text{ice}}$ between 1-10 L$^{-1}$. Measurements from the CDP showed $N_{\text{drop}} \sim 200$ cm$^{-3}$ and size distributions from the same instrument at 08 CET revealed generally small droplets of mode size 12 µm (fig. 11b). Ice fractions during this time (08 CET) were generally < 0.2 despite some occasional variation due to changes in cloud microphysical structure. From 09 CET $N_{\text{ice}}$ and IWCs gradually increased over an approximately 8 hour period to near 400 L$^{-1}$ and 2 g m$^{-3}$ respectively by 17 CET with peaks up to 800 L$^{-1}$. Ice fractions during the period varied between ~ 0.6 and 1. Ice crystals in this region of highest mass and concentration values were generally small. A 5 minute size distribution from the 2D-S at 17 CET (fig. 11a) showed mode diameters centred around 90 µm and 300 µm. Comparing this size distribution with one from the start of the period (fig. 11a 08 CET), where $N_{\text{ice}}$ values were relatively low, shows the cause of the increase in concentrations is similar to case 1, where particle numbers are enhanced across the measurement range of
the instrument. CDP size distributions at the same time (fig. 11b) showed a broadening of the droplet distribution compared to the earlier period where ice fractions were low. Over the entire transition period between 06 CET 30 Jan and 00 CET 31 Jan, despite \( N_{\text{drop}} \) gradually decreasing with a corresponding trend for increased \( N_{\text{ice}} \), there is variation in LWC and IWC that leads to short term reversals of the trend. This process is represented by occasions in which there are rapid falls in LWC. For example at 11 CET there is a fall in LWCs to generally < 0.1 g m\(^{-3}\) and an increase in IWCs to ~ 0.4 g m\(^{-3}\). This change led to many ice fraction values equal to 1 (completely glaciated cloud). At ~ 22 CET the pattern of predominantly high ice fractions was reversed as LWC rose to 0.3 g m\(^{-3}\) and \( N_{\text{drop}} \) increased to 150 cm\(^{-3}\) as \( N_{\text{ice}} \) fell to a few 10s per litre. Ice fractions responded with values generally decreasing to < 0.2 by 23 CET.

During the second period between 06 CET and 09 CET 31 January the ice fraction of the cloud increased from < 0.2 to > 0.8 and then the pattern repeated itself. Each cycle lasted approximately 1 hour. During the first cycle droplet number concentrations remained stable, around 100 cm\(^{-3}\), with peaks up to 200 cm\(^{-3}\) and LWCs of approximately 0.1 g m\(^{-3}\). Before ice fractions increased, the CDP size distributions, around 0620 CET, were bimodal, with modes centred around 5 and 15 \( \mu \)m (figure 11b). 2D-S imagery from this period showed some irregular ice crystals together with small droplets (fig. 12c).

In the region with highest calculated ice fractions (0655 CET) 2D-S size distributions showed bimodality with a dominant mode around 80 \( \mu \)m and a larger mode at 230 \( \mu \)m (Figure 11a). 2D-S images during this period revealed an increase in ice crystals, but small liquid droplets persisted (fig. 12d). The major cause of the change in ice fraction was due to two gradual increases and then decreases in \( N_{\text{ice}} \) from concentrations of a few 10s per litre, during low ice fraction periods, to ~ 100 L\(^{-1}\) when ice fractions were relatively high. Despite some variability in the liquid phase, \( N_{\text{drop}} \) and LWC during these periods tended to remain fairly constant around 100 cm\(^{-3}\) and 0.1 g m\(^{-3}\) respectively. Temperatures over the period rose approximately 1 °C to -12 °C and wind speed increased from near 6 m s\(^{-1}\) to 9 m s\(^{-1}\). The wind direction showed little variability and came from the southerly sector throughout.

### 5.3 Saharan Dust Event (Case 3)

Over the period from 30 January to 3 February Switzerland experienced a Saharan Dust Event (SDE) and this was measured at the Schilthorn and JFJ sites. The wind direction throughout this period was southerly and back trajectories showed the airmass origin to be split between Europe and Africa. Trajectories from Africa originated from a low altitude,
consistent with the transport of Saharan dust to JFJ. Temperatures showed significant change from near -20 °C at the beginning of the period before increasing to ~ -10 °C at 12 CET on 2 Feb. At Schilthorn the GRIMM OPC measured dust concentrations that peaked at 18 CET on 2 Feb up to ~ 3000 L⁻¹ over the size range 0.5 < \(D_p\) < 10 µm. The WIBS4, measuring over the size range 0.8 < \(D_p\) < 20 µm, measured peak dust concentrations (~ 2500 L⁻¹) approximately 12 hours earlier in the day. The WIBS4 has the ability to distinguish between UV-fluorescent and non UV-fluorescent particles, e.g. Robinson et al. (2013). The numbers of particles found to fluoresce was approximately 100 L⁻¹ during the main SDE peak (fig. 13). We analysed size distributions from both the WIBS4 (JFJ) and the GRIMM OPC (Schilthorn) during the SDE and for periods outside the SDE (fig. 15). Outside of the dust event aerosol particle size was generally limited to < 10 µm, however during the SDE we observed a broadening of the size distribution. We found concentrations of smaller mode aerosol particles (< ~ 1 µm) remained similar within and outside the SDE, with the main enhancement of concentrations during the SDE being due to increased numbers of particles > 1µm. When examining the size distributions of UV-fluorescent particles outside of and during the SDE we also found an increase in the mode size of particles to increase from ~ 1 µm to 2 µm. Over the SDE period there was also a notable broadening of the fluorescent particle size distributions (Fig. 15).

For this measurement period, between 30 January and 3 February 2014, we examined the link between aerosol concentrations (UV-fluorescent and non UV-fluorescent) at Schilthorn and \(N_{ice}\). Analysis of wind speed and direction at JFJ and Schilthorn showed that the two sites were connected throughout this measurement period. Figure 14 shows scatter plots of \(N_{ice}\) (coloured as a function of temperature) measured at JFJ by the 2D-S plotted against the concentration of fluorescent aerosol measured by WIBS4. In this paper we generally removed aerosol data for periods when the relative humidity was > 80 % for this site. However, due to the limited period over which this SDE occurred we have relaxed these criteria to include aerosol measurements that were made at relative humidities as high as 91 %. Fig. 14a shows a peak in ice concentrations around -15 °C that is independent of the UV-fluorescent aerosol concentrations, however, when examining a subset of the data, where ice concentrations are < 100 L⁻¹ (Figure 14b,c) we see that there is some agreement between the number of UV-fluorescent aerosol particles measured by the WIBS4 at Schilthorn and the number of ice crystals observed at JFJ. The coloured markers show that this association appears to apply for relatively high temperatures around the -10 °C range. If we isolate data points to exclude measurements at temperatures below
- 15 °C (fig. 14b) and -12 °C (fig. 14c) we see an increasing correlation between \( N_{\text{ice}} \) and the concentration of UV-fluorescent aerosols.

5.4 8 February 2014 (Case 4)

Between 1130 CET and 1230 CET on 8 February, fluctuations in the ice fraction between 0 and 1 were associated with very swift transitions between glaciated, liquid and mixed phase conditions. Figure 16 shows that period in which ice fractions oscillate between values of 1 (glaciated), and 0 (liquid). There are also periods where ice fractions lie between these values, representing mixed phase microphysics. These fluctuations between glaciated and liquid conditions were observed to happen quickly – often over 1 second time periods. For this reason the microphysics data presented in figure 16 is displayed as 1 Hz averaged data to better show these changes. Generally peaks in \( N_{\text{drop}} \) and \( N_{\text{ice}} \) were observed to be a few tens per litre with swift changes between the two. Peaks in the IWCs and LWCs were around 0.1 and 0.02 g m\(^{-3}\) respectively. Shadow imagery from the 2D-S probe (Fig. 12a,b) highlight pockets of glaciated cloud together with small liquid droplets. During the measurement period the wind direction was from the southerly sector throughout, with some variation between 100° and 170° wind angles. Temperatures were generally between -10 °C and -15 °C, but showed frequent variation of magnitude in the order of 4 °C.

6.0 Ice Nuclei Predictions

At the Schilthorn site measurements of aerosol size distributions provided by the GRIMM instrument were used as input to the primary ice nucleation parameterization developed by DeMott et al. (2010) (hereafter referred to as \( D10 \)). This scheme used data collected from 9 measurement campaigns of IN using a continuous flow diffusion chamber, over a 14 year period of field campaigns at a variety of locations over the temperature range -9°C to -36°C and where saturation with respect to water was > 100%. An additional constraint was to use data only where aerosol size distributions were also available. They provided a parameterization based on both the number of aerosol particles > 0.5 \( \mu \text{m} \) diameter and temperature. This parameterization predicts 62% of the observed IN to within a factor of 2 (\( D10 \)), although they highlighted the need for further geospatial data sets to test and improve this parameterisation.
Aerosol data for use with *D10* was selected during periods in which relative humidity (RH) was < 80% to reduce artefacts in the data caused by sampling during in-cloud periods. The maximum particle diameter measured in *D10* was 1.6 µm and for this reason we also limit particle concentrations to the size range $0.5 < D < 1.6$ µm for input into the scheme. Here we use the parameterization (Eq. 2) to predict IN concentrations at JFJ based on aerosol properties at Schilthorn. The parameterization is represented as:

$$n_{\text{IN},T_k} = a(273.16 - T_k)^b(n_{\text{aer},0.5})^{c(273.16 - T_k) + d}$$

Where $a = 0.0000594$, $b = 3.33$, $c = 0.0264$, $d = 0.0033$, $T_k$ is the cloud temperature in degrees Kelvin, $n_{\text{aer},0.5}$ is the number concentration of aerosol particles $> 0.5$ µm and $n_{\text{IN},T_k}$ is the ice nuclei number concentration ($\text{L}^{-1}$) at $T_k$.

The Schilthorn and JFJ sites are approximately 11 km apart at altitudes of 3580 m and 2970 m asl respectively. Changes in wind speed at each site showed a generally good correlation and based on this we have assumed both to be influenced by the same airmass. When we compared ice crystal concentrations, $N_{\text{ice}}$, observed at JFJ (fig. 17) with the *D10* prediction we found that they were underpredicted by as much as 2 to 3 orders of magnitude. Figure 17 shows the concentration of aerosols $> 0.5$ µm in size versus $N_{\text{ice}}$. If we assume that a greater number of these particles are acting as IN than the *D10* scheme predicts then this could explain the ice concentrations we observed at JFJ. There is however no strong correlation between this concentration and ice crystal number.

### 7.0 Discussion

Measurements of cloud microphysical structure at JFJ during January and February 2014 revealed constant variation in the liquid and ice properties of the cloud. Synoptic conditions at the site led to significant periods of strong southerly winds as depressions stalled across Western Europe. Wind data from the site showed that the origin to be almost exclusively from either a northerly (315°-45°) or south easterly (115°-165°). Schilthorn exhibited a different prevailing wind direction, which was mostly from the south west. Despite this back trajectories showed that both sites were influenced by the same airmass origins and for this reason we assumed aerosol measurements made out of cloud at the Schilthorn to be broadly representative of those at JFJ. However it is possible the complex air flows in this high alpine region could have led to some degree of recirculation of the airmass that could impact on local aerosol measurements.
Analysis of ice and liquid properties from the 2D-S and CDP probes showed cloud microphysical structure to exhibit large amounts of variability in the absence of any significant change in ambient conditions such as wind speed, direction and temperature. This variability was very often represented by changes in the ice mass fraction. The cases described here were selected due to evidence of these changes in the relative ice and liquid mass values. However the scale of these changes was observed to differ between case studies. For example on the 11 February 2014 (Case 1)(Fig. 6,7) measurements showed two transitions highlighted by a change from high to low ice fraction values over a period of approximately 8 hours. In another instance on 30 January 2014 (Case 2) ice fractions were observed to be generally low (< 0.2), before increasing to values > ~ 0.8 and remaining stable for about 12 hours prior to decreasing again to values < 0.2. On 31 January 2014 during Case 2 there was evidence of a cyclical pattern to ice fraction values, with a repeating pattern of increasing and decreasing ice fraction values over a time period of ~ 1 hour. During case 4 ice fractions were highly variable with rapid changes over short time periods of approximately 1 second, representing rapid changes between liquid, mixed phase and glaciated cloud. Images from the 2D-S and CPI probes showed regions of glaciated cloud amongst liquid droplets. Contributions to changes in the ice fraction values were seen to be due to a several different factors associated with changing ice and liquid mass values. A number of different scenarios existed by which changes in the mass of ice and liquid phases led to changes in the sign and magnitude of the ice fraction. For example during the campaign period we observed increasing ice fractions due to:

1. Constant liquid water content and increasing ice water content
2. Decreasing liquid water content and increasing ice water content
3. Decreasing liquid water content and constant ice water content
4. Increase in ice water content is greater than increase in liquid water content

and periods of decreasing ice fraction were associated with:

1. Constant ice water content and increasing liquid water content
2. Increasing liquid water content and decreasing ice water content
3. Increasing liquid water content and constant ice water content
4. Increase in liquid water content is greater than increase in ice water content

It is important to recognise the range of conditions that lead to changes in the ice fraction due to the implications for the processes driving any particular transition. Studies have shown that the availability of IN at a given temperature range is related to the size (DeMott et al., 2010) and composition (Hoose et al., 2012) of an aerosol particle. Changes in ice
number concentrations at JFJ (and therefore ice fractions) over the temperature range \(-10 \, ^\circ\text{C} < T < -36 \, ^\circ\text{C}\) in the absence of influence from homogeneous and secondary freezing processes are likely to be attributed to variability in the number of heterogeneous freezing nuclei carried to JFJ.

Increasing ice fractions in this study were often due to the presence of high \(N_{\text{ice}}\) that was sometimes in excess of 2000 L\(^{-1}\) at temperatures \(-15^\circ\text{C}\). During all the cases described we found that the the \(D10\) parameterization could not explain the high \(N_{\text{ice}}\) observed at JFJ, and generally under predicted ice concentrations by up to several orders of magnitude.

On 1 February 2014 a Saharan dust event (Case 4) was sampled at JFJ and Schilthorn. We have found a possible association between the number of UV-fluorescent aerosol particles measured at the Schilthorn site and the concentration of ice particles at JFJ. During this period temperatures varied between approximately \(-9^\circ\text{C} < T < -20^\circ\text{C}\) and scatter plots (Fig. 14) of \(N_{\text{ice}}\) at JFJ vs the UV- fluorescent \((N_{\text{FL}})\) particle concentrations measured by the WIBS at Schilthorn showed high concentrations that appeared to be independent of the FL concentration around \(-15 \, ^\circ\text{C}\) but if we consider the temperature range up to \(-10 \, ^\circ\text{C}\) the data suggests a possible correlation between \(N_{\text{ice}}\) and \(N_{\text{FL}}\). It is clear that when we isolate data points that represent measurements taken at higher temperatures (as in figure 14 a and b) we see better agreement. These aerosol are likely to represent biological particles that are known to act as efficient ice nuclei in the temperature range warmer than \(-15 \, ^\circ\text{C}\). These can include particles such as bacteria (Lindow, 1983) and pollens (Diehl et al., 2001; Diehl et al., 2002) while studies have shown the impact these can have on clouds and precipitation processes in the natural atmosphere (Creamean et al., 2013).

Laboratory experiments using the WIBS4 instrument (Toprak & Schnaiter, 2013), suggest that the maximum mis-classification of mineral dusts as biological particles, due to naturally UV-fluorescent rare-earth activant content, is typically of the order of 10%. Similar experiments with Saharan dust samples by us, (Gabey, 2010), suggests mis-classification percentages of 6.8%. Other possible mis-classification due e.g. soot particles is estimated to be 1 particle per litre under typical polluted conditions, (Toprak & Schnaiter, 2013). It should be noted however that these studies did not investigate whether some of the UV-Fluorescent behaviour associated with the mineral particles was in fact due to biological material on their surface. Laboratory studies of collected SE Asian dusts, e.g. Yamaguchi (2013), have shown significant UV-fluorescent surface contamination associated with micro-organisms. Within the SDE, UV-fluorescent particle concentrations reached levels of 100 L\(^{-1}\), with mode sizes of 2 µm. This compares with typically \(~10\) L\(^{-1}\)
and 1 µm outside the event. Using a conservative miss-classification as biological of 10-15%, it is possible that the fluorescent particles measured by the WIBS could be artefacts. However the finding of a correlation between these particles and concentrations of ice at JFJ suggests they were likely to be of biological origin an IN active in temperatures around -10 °C. At colder temperatures < -15 °C we did not observe a link between $N_{FL}$ and $N_{ice}$ at JFJ. At temperatures below - 15 °C other aerosols dominate heterogeneous freezing, such as dust (Ansmann et al., 2005) and carbonaceous particles such as soot. However when comparing ice concentrations non-fluorescent aerosol concentrations, likely to represent dust from the Sahara Desert we did not find a correlation in the temperature range these particles are likely to act as IN. Size distributions (Fig. 8 and 11) showed a persistent mode of ice crystals around 80 µm, with some contribution from larger particles that were several hundred microns in diameter. The generally small size of these crystals suggests they were produced locally and in many cases may have existed for about a minute before being measured (assuming they are in a water saturated environment). When examining differences between cloud containing low and high ice fractions we found that the differences in $N_{ice}$ were due to changes in concentrations of ice crystals across most of the size spectrum, rather than any contribution from a limited size range.

During the measurement period two dominant wind directions were evident and when analysing ice concentrations as a function of wind direction we found the highest concentrations to be associated with periods when the wind originated from the northerly sector. The source of this bias is unclear but it is appropriate to consider the location and unique topography surrounding the measurement site. To the south the Aletschgletscher descends slowly 23 km down the valley and to the North another Glacier involves a near vertical drop to the ski area of Kleine Scheidegg. The mountain peaks of Jungfrau (4158 m asl) and Mönch (4107 m asl) rise above the measurement site. During most of the year the surface around JFJ is covered with snow and research has shown that this can be lifted by the wind into the air above the surface. These particles are generally small (< 200 µm) and can be blown in high concentrations > ~ 100 L$^{-1}$ up to a metre above the surface depending on ambient conditions such as wind speed, temperature, relative humidity and whether there has been any recent snowfall (Gordon and Taylor, 2008). This process undoubtedly takes place on the surfaces surrounding JFJ, but understanding the impact on cloud microphysics datasets is complex. When looking at background particle concentrations during clear sunny days at JFJ we found the contribution from blowing snow particles to be negligible. Generally concentrations were < 10 L$^{-1}$ but very often no particles were measured. However the process is complex and during clear days with low relative
humidity the ice crystals lifted from the surface may sublime. When cloud covers JFJ and the surrounding surfaces the physical processes are very different. Ice particles stripped from the surface by wind are added to a supersaturated environment with respect to ice and these small particles grow quickly through vapour diffusion. It is possible significant concentrations of blown snow particles enter the cloud and grow rapidly, eventually exhibiting the growth habit of the temperature regime at that given time. These particles would likely be measured by the cloud probes and it would not be possible to remove them from this data set.

Another possible source of enhanced ice concentrations from the surface could be due to the availability of 'preactivated' aerosol particles. These are particles that have already acted as an IN, been scavenged by growing ice crystals or were simply exposed to a temperature lower than 235 K. Wagner et al. (2014) investigated aerosol particles before and after pre activation at the AIDA cloud chamber at the Karlsruhe Institute of Technology (KIT) and showed that the IN efficiency of these pre activated particles is increased. Ice on the surface of the surrounding mountains and glaciers at JFJ will sublime in conditions when saturation with respect to ice is < 1. This may lead to the presence of pre-activated particles on the surface and it is possible that these are blown into the cloud above the surface, leading to an increased IN population, enhancing the number of ice particles observed.

The surface as a source of ice is likely to play a role at the high alpine site JFJ, but understanding why this would have greater impact when the wind is from the north is unclear. Another possible influence on measurements at this site is related to land use close to JFJ. To the South there is very little human activity, but to the North operations at the Jungfrau ski area to produce artificial snow may provide a source of IN. The use of IN active at high temperatures in snowmax, (pseudomonas syringae bacteria used to generate artificial snow and IN active between -5 and -10°C), may introduce higher concentrations of IN to the clouds just north of JFJ that seed the clouds that are eventually measured at the site. This may be one possible explanation of the finding that higher ice concentrations were generally observed when the wind was from a northerly direction.

High $N_{ice}$ in clouds in certain conditions is often associated with secondary ice processes. The most influential of these appears to be the Hallett-Mossop (HM) process, (Hallett and Mossop, 1974). This process leads to enhancement of $N_{ice}$ through rime splintering but is restricted to the temperature range $-3 \ ^{\circ}C < T < -8 \ ^{\circ}C$. The clouds observed at JFJ over the measurement period were generally colder than this temperature zone and for this reason
we do not expect the HM process to have made any significant contribution to the $N_{ice}$ concentrations that we observed. There is some evidence (see for example Pruppacher and Klett, 1997) that ice crystal multiplication can also occur by the shattering of liquid drops as they freeze, by the fragmentation of ice particles either on collision with other particles or as they evaporate. The proposed mechanism for the production of splinters on the freezing of a supercooled drop is that in the first stage of freezing ice is produced in the body of the droplet as it warms up to 0 °C. This then nucleates an ice shell, which spreads round the drop as heat is lost to the environment. As the drop freezes further with a shell of ice the pressure in the drop rises and disruption of the shell occurs producing protuberances, the ejection of gas bubbles, supercooled water and ice fragments. It has been observed in laboratory studies that the drop may fragment into 2 or more parts on freezing. The contribution of this process to ice multiplication in clouds is poorly quantified, however there is little evidence to suggest that this is a major source of secondary ice. For example Rango (2008) found evidence that this process operated in shallow marine frontal clouds producing a modest number of ice particles consistent with laboratory studies but the process is not sufficiently powerful to explain a large enhancement of ice crystal concentrations such as observed in this paper.

Another process often reported in the literature is the fragmentation of ice crystals particularly delicate shapes such as stellar or dendritic crystals (see for example Griggs and Choularton, 1986). There is some evidence of fragmentation in natural clouds (e.g. Hobbs et al. 1980), however many of the early studies that reported evidence of fragments around -15 °C (e.g. Bower et al., 1996) were possibly contaminated by the fragmentation of ice crystals on the inlets of airborne probes (Field, Heymsfield, & Bansemer, 2006). Recent airborne studies in natural clouds have found little evidence for this breakup process e.g. Lloyd et al. (2014a,b) and Crosier et al. (2011) in layer clouds and Crawford et al. (2012) in convective clouds. However Yano et al. (2013) have published calculations to suggest that this process may be important in clouds where rime-splintering by the Hallett-Mossop process is not significant. In view of the results presented in this paper this process cannot be ruled out and further investigation is needed.

8.0 Conclusions

Observations of cloud microphysical structures and their association with aerosol properties have been presented from a measurement campaign that took place at Jungfraujoch and Schilthorn, Switzerland.
• Changes in the LWC and IWC lead to significant changes in ice mass fraction values that take place over temporal scales of second to hours.

• Ice fraction values are influenced by different combinations of changes in the liquid and ice water contents measured at Jungfraujoch.

• Ice number concentrations varied from zero during liquid cloud periods to over 2000 L\(^{-1}\) during mixed phase and glaciated cloud periods.

• Ice crystals were found to be generally small (< 300 µm), with bi-modality sometimes evident in the size distribution with a mode centred around 80 µm and another at 200 µm.

• Using aerosol measurements from the Schilthorn site as input to the DeMott et al. (2010) primary ice nucleation scheme we could not account for the high concentrations of ice particles observed at Jungfraujoch.

• It is possible the surface was a source of pre activated aerosol and blown snow, which could enhance ice concentrations.

• It is unclear why we observed higher concentrations of ice when the wind was from a northerly direction. This may be due to a land use impact as the Jungfrau ski area uses snow cannons to produce artificial snow, providing a potential additional source of IN just to the north of Jungfraujoch.

• During the campaign a significant Saharan Dust Event was detected and a possible link was found between the concentration of UV-fluorescent aerosol and ice crystal concentrations at Jungfraujoch at relatively high temperatures, around -10 °C, but not with the dust particles at lower temperatures.
Figures

Fig. 1. Satellite imagery showing the locations (red boxes) of Jungfraujoch (A), Schilthorn (B) and Kleine Scheidegg (C). Source: Google Earth.

Fig. 2. Photograph of the instruments mounted on a rotator on the platform at the Sphinx Laboratory. Manchester instruments include a Metek for measuring wind speed, 3V-CPI for cloud particle imagery, CDP for droplet measurements, CIP-15 for cloud particle imagery, CAS for droplet and aerosol measurements and an FSSP for droplet
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Fig. 4. Wind rose showing wind direction frequency (%) and wind velocity (m s\(^{-1}\)) for Schilthorn (a) and Jungfraujoch (b).

Fig. 5. Time series data from the meteorological station maintained by Meteo Swiss at the Jungfraujoch site for Relative Humidity (%) (purple trace), Temperature (°C) (green trace), Wind Velocity (m s\(^{-1}\)) (black trace) and Pressure (mb) (red trace).
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Fig. 8. Size distributions during ice fraction transition period one and two from case 1 on 11 and 12 February for ice particles measured by the 2D-S (a) and liquid droplets measured by the CDP (b).
**Fig. 9.** Time series from ice fraction transition one on 30 January 2014 for ice number concentration from the 2D-S (L⁻¹) (red trace), liquid droplet concentration from the CDP (cm⁻³) (blue trace), liquid water content (LWC) from the CDP (g m⁻³) (green trace) and ice water content (IWC) from the 2D-S (g m⁻³) (purple trace), temperature (°C) (purple trace), wind speed (m s⁻¹) (brown trace) and ice fraction (black trace). Labels A and B mark 5 minute 2D-S and CDP size distributions from 0800 CET and 1700 CET 11 Feb 2014.
**Fig. 10** Time series from ice fraction transition two on 30 January 2014 for ice number concentration from the 2D-S (L$^{-1}$) (red trace), liquid droplet concentration from the CDP (cm$^{-3}$) (blue trace), liquid water content (LWC) from the CDP (g m$^{-3}$) (green trace) and ice water content (IWC) from the 2D-S (g m$^{-3}$) (purple trace), temperature (°C) (purple trace), wind speed (m s$^{-1}$) (brown trace), wind angle (°) (grey trace) and ice fraction (black trace).

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Fig. 12. Examples of 2D-S Imagery showing swift transitions (a and b) between liquid, mixed phase and glaciated cloud on 8 February 2014 and particle imagery from a period of low ice fractions (c) and high ice fractions (d) on 31 January 2014.

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Fig. 17. Ice concentrations measured by the 2D-S at Jungfraujoch plotted against predicted ice nuclei values from the DeMott et al. (2010) primary ice nucleation parameterization (grey markers) and total aerosol particles > 0.5 µm measured by the GRIMM at Schilthorn (green markers). Diagrams a, b and c refer to 30-31 January (case 2), 8 February (case 4) and 11-12 February (case 1) respectively.
<table>
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<th>Measurement</th>
<th>Method</th>
<th>Size range [µm]</th>
<th>Time Resolution</th>
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<td><strong>2DS</strong>: Particle size distributions and shadow imagery <strong>CPI</strong>: Particle size distributions and particle photographs</td>
<td><strong>2DS</strong>: Optical Array Probe (128 element array at 10 µm effective resolution) <strong>CPI</strong>: Use of a CMOS(or CCD) Camera to photograph particles</td>
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**Table 1:** A summary of the instrumentation deployed at the Jungfraujoch site including information about measurement type, method, size ranges and time resolution of the data provided.
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**Table 2:** A summary of the instrumentation deployed at the Jungfraujoch site including information about measurement type, method, size ranges and time resolution of the data provided.

**Bibliography**


Gabey, A.: Laboratory and field characterisation of fluorescent and primary biological aerosol particles, University of Manchester, 2011.


Lloyd, G., Dearden, C., Choularton, T. W., Crosier, J., & Bower, K. N.: Observations of the origin and distribution of ice in cold, warm and occluded frontal systems during the DIAMET campaign. Monthly Weather Review, American Meteorological Society. (Accepted)


8.1 An Overview of the Work Presented in this Thesis

This thesis focuses on observations of cloud microphysics through in-situ measurements during both airborne and ground based campaigns and remote sensing in one case during the DIAMET campaign. There was a particular focus on identifying the processes that led to the formation and distribution of ice within the clouds studied and the role of secondary ice in producing high concentrations of ice in the H-M temperature zone. Each case presented the opportunity to measure cloud microphysical structure with modern cloud probes, allowing us to characterize the dominant microphysics in each instance and to observe any significant transitions. This thesis included work from the DIAMET, ACCACIA and INUPIAQ campaigns, each with their own specific set of aims (as discussed in the relevant chapters). In each project accurate and detailed measurements of cloud microphysical properties was a priority and one of the main motivations for this work. The measurements were then used in a number of ways to advance our knowledge of the cloud processes in each study, which included: reporting our observations and comparing and contrasting them with previous measurements; assessing the impact of our observations on the cloud structure; addressing the suitability of a primary ice parameterization to predict our observed ice number concentrations and to look in detail at the role of secondary ice processes in each case and the microphysics in which it was associated with. The main findings from each paper presented in this thesis is now outlined below.

8.1.1 Paper 1 - DIAMET

The first paper (Ch. 4) presented microphysics observations using a suite of instruments to make in-situ measurements within warm, cold and occluded frontal systems during the DIAMET campaign. The microphysical structure and mesoscale characteristics of each
case were discussed and contrasted with one another. The main findings are summarised below.

- The formation of ice in each case was found to be different. The warm and occluded frontal cases consisted of high and cold cloud tops < -37°C, which led to homogeneous ice formation. The cold front case was characterised by a kata cold front consisting of a narrow cold frontal rainband (NCFR) that had lower altitude and warmer (~-13°C) cloud tops. In this case the origin of the ice was very likely to be through primary heterogeneous ice nucleation on available ice nuclei. The number of ice particles observed was broadly consistent with predictions from the DeMott et al. (2010) primary ice paramaterization.

- The relatively warm cloud tops of the cold frontal system were in close proximity to the H-M temperature zone below and these two areas became coupled with the ability to directly influence each other.

- In all three frontal systems the highest ice number concentrations and ice water contents were observed in the H-M temperature zone, where secondary ice production via rime-splintering enhanced concentrations of ice particles by about an order of magnitude.

- All three cases were observed to contain large supercooled droplets > 80 µm that were found to be in close proximity to high concentrations of ice formed through the H-M process. These larger drops could play an important role in 'short cutting' the process. This is because upon freezing they become instant rimers with significant surface area that increases the rime accretion rate - leading to greater splinter production.

- The dynamics of the Kata cold front had a direct impact on the spatial distribution of precipitation and on the positioning of the SIP zone. This happened as a component of forward motion aloft, associated with these types of fronts, pushed ice crystals nucleated close to cloud top ahead of the surface front. Vertical motions were inferred using data from the 3GHz meteorological radar at CFARR and the movement of developing ice crystals observed through analysis of differential reflectivity fields.

- In all cases heavily rimed snow particles were observed and these are very likely to have been the sites on which rime-splintering took place.
The instantaneous rates of latent heating and cooling for liquid and ice phase processes associated with the narrow cold frontal rain band were quantified using a combination of the in-situ, measurements from the observational microphysics paper, and a Lagrangian parcel model framework. We focused on in-situ measurements taken during the final stages of a cold frontal transition from ana to kata type. It was shown that that the diabatic terms of largest magnitude are associated with the liquid phase, i.e. from condensation and evaporation, in line with previous idealized modelling studies. The diabatic effects of ice phase processes (deposition/sublimation and melting) were found to be at least an order of magnitude smaller than the peak heating rates from condensation. The diabatic effects of riming were an order of magnitude smaller still in this case. The diabatic effects of the ice phase covered a wide horizontal extent, and in the case of melting, were concentrated within a shallow layer.

Precipitating ice is shown to produce a cooling effect via sublimation and melting which could influence the mesoscale evolution of the front by modifying the temperature profile below cloud base. Based on this case study, we conclude that in terms of PV generation, the role of the ice phase is most likely to be important during the ana phase of the front, where sublimation and melting can directly influence the strength of the cold air inflow. Then as the system matures and the front transitions from ana to kata, the diabatic influence of the ice phase is reduced as a result of the dry intrusion aloft, which pushes the ice into the warm sector ahead of the cold front.

In this paper we also attempted to validate the performance of a typical bulk microphysics scheme in terms of its representation of diabatic heating and cooling, using a detailed bin-resolved scheme and the in-situ observations as the benchmark. Our main conclusions from this comparison are: For the conditions encountered in this study, the method of saturation adjustment used to represent condensation growth in bulk schemes was found to be an excellent approximation when compared to explicit treatments. Bulk schemes that treat ice crystals as simple spheres are shown to be inadequate in terms of simulating local rates of diabatic heating and cooling associated with changes in ice crystal mass, particularly via vapour diffusion and melting, calling for bulk schemes to be able to account for variations in ice crystal shape in order to simulate more realistic diabatic heating and cooling profiles. Ventilation effects were found to influence the magnitude of the local diabatic heating and cooling rates by as much as a factor of two or more. The bulk schemes do not include the effects of crystal habit and therefore tend to overestimate the effects of ventilation. The use of negative exponential functions to describe the form of ice
particle size distributions in the bulk scheme was generally found to represent an adequate fit to the observed data.

8.1.2 Paper 2 - ACCACIA

The second paper (Ch. 6) described microphysics measurements made around the archipelago of Svalbard during spring and summer 2013 during the ACCACIA project. The microphysics of arctic stratocumulus were investigated and the results compared and contrasted with each other and those from previous studies that have taken place in the Arctic and Antarctic. The main findings are summarised below.

- Two cases from both the spring and summer campaign were presented. The cloud layers in the spring spanned a colder temperature range (~ -10 °C ≥ T > -20 °C) than in the summer cases (generally 0 °C ≥ T > -10 °C).
- One of the spring cases was found to have significantly higher aerosol concentrations (~ 300-400 cm⁻³) compared to the other (~ 50-100 cm⁻³). However ice concentrations in both cases were similar, suggesting the source of increased aerosol was not providing an increase in ice nuclei over the temperature range the cloud spanned.
- In the spring cases clouds with approximately adiabatic LWCs precipitated low concentrations (generally < 1 L⁻¹) of ice from liquid cloud tops. These particles grew to median diameter ~ 500 µm with the dominant growth habit found to be dendritic in nature.
- In the summer cases liquid cloud tops were still evident and were seen to precipitate ice into lower layers. However the higher temperatures that these clouds spanned led to secondary ice production through the H-M process, enhancing ice concentrations by about a factor of 5 (or more) above the concentrations observed in the spring cases.
- The observations suggest that summer Arctic stratocumulus may contain higher ice number concentrations than similar clouds in the cold season.
- When using the primary ice nuclei parameterization of DeMott et al. (2010) to predict ice concentrations, using aerosol measured during the flights as input, we found the scheme tended to over predict our median values by up to a factor of 4, but under predicted when compared to our peak values. During the summer cases,
due to contributions from secondary ice production, the scheme predicted significantly lower concentrations of ice particles.

- Observations of peak ice crystal concentrations above the primary IN prediction from D10 could possibly be evidence for secondary ice production through fragmentation of the frail ice crystal shapes we observed during ACCACIA
- When comparing our findings with those from the Antarctic (Grosvenor et al., 2012) we found approximately an order of magnitude higher primary ice concentrations and higher concentrations of aerosol particles > 0.5 µm. They also observed enhancement of ice concentrations through secondary ice production in the warmer clouds that they observed, but the concentrations were about an order of magnitude less than our observations in the Arctic.

8.1.3 Paper 3 - INUPIAQ

The third paper (Ch. 7) examined microphysics measurements made during a multi-site campaign that took place as part of INUPIAQ. The characteristics of the microphysics and their association with aerosol concentrations and composition was reported from this ground based campaign. The measurements revealed:

- Frequent, significant changes in ice mass fractions due to transitions in the number of ice and liquid particles that took place on temporal scales of just seconds to hours.
- Changes in the ice fraction values could be attributed to a number of different processes that caused changes in the cloud microphysics at Jungfraujoch.
- Large variation was observed in ice number concentrations, with times of liquid dominated cloud, and other periods where ice number concentrations reached over 2000 L$^{-1}$.
- Ice crystals were found to be generally small (< 300 µm) with bi-modality sometimes evident in the size distributions, with modes centred around 80 µm and 200 µm.
- We used the primary IN parameterization of DeMott et al. (2010) with aerosol measurements made at Schilthorn. This site is approximately 500 m lower and measures aerosol entering the cloud when the wind flow is from the north, or leaving the cloud with flow from the southerly sector. We could not account for the
high concentrations of ice at JFJ, with the scheme underestimating ice nuclei concentrations by up to 3 orders of magnitude.

- When considering the surface as a potential source of the high ice concentrations observed we found that it may be possible that wind-blown snow and pre-activated aerosol particles could be increasing cloud ice concentrations at JFJ. In addition the impact of human activity at the Jungfrau ski area to the north could introduce a source of ice nuclei through the use of snow cannons to produce artificial snow.

- During the campaign period we observed a Saharan dust event and a possible link was found between the concentration of UV-fluorescent particles measured at Schilthorn and the number of ice particles measured at JFJ in the temperature range ~ -10 °C. The fluorescent particles were likely to have been of biological origin. When examining the link between ice crystal number concentrations and non-fluorescent particles measured at Schilthorn no association was found.

8.2 Discussion and Future Work

One of the main outcomes of this work was the finding that enhancement of ice number concentrations in the H-M temperature zone, due to rime-splintering, was very common in a variety of different situations that included the three frontal system types during DIAMET and arctic stratocumulus cloud in the ACCACIA project. The process is clearly relevant to many different cloud systems in the atmosphere and has the ability to dominate the ice phase of a cloud in terms of mass and number concentrations. Classically, the H-M process occurs as the riming process takes place on graupel particles and many studies (see Chapter 2.3) have investigated the splinter production rate per milligram of rime based on riming velocities applicable to the fallspeed of graupel particles. However it is clear that snow particles, with lower fallspeeds, were likely to be acting as riming sites in the cases presented here. Saunders and Hosseini (2001) conducted the most extensive study into splinter production as a function of velocity over the range 1.5 - 12 m s⁻¹, but experimental verification of splinter production at low riming velocities (similar to that of snowflakes) will be an important step in better quantifying the process. The initiation of the ice phase is important for the development of precipitation as ice particles gain mass through the Bergeron-Findeisen process before sedimenting out of the cloud. However the exact impact of high concentrations of ice, for example those produced through secondary processes, on precipitation is uncertain.
In the cases where secondary ice splinters were encountered we often observed large liquid droplets in close proximity. Modelling studies (e.g. Crawford et al. 2011) have shown that these larger droplets are important for triggering glaciation of a cloud through SIP. Although we could not directly link the high concentrations of ice we observed in our cases to the presence of the larger liquid droplets it is possible that these are very important in speeding up the process. Future microphysics measurements that encounter the H-M temperature zone should aim to identify any relationship between SIP and the existence of larger liquid droplets. If these larger droplets freeze they become instant rimers, and provide a powerful mechanism by which large numbers of splinters are produced very rapidly.

There has been some focus on phase transitions in Arctic clouds and the temperature range in which this may occur. However, this has been in relation to primary heterogeneous ice nucleation on available ice nuclei. We observed secondary ice to play an important role in the glaciation of summertime arctic clouds in the H-M temperature zone, which served to increase ice concentrations above what was observed in arctic stratocumulus clouds spanning colder temperatures. A negative correlation between temperature and ice concentrations has been observed before (Hobbs and Rangno, 1998) and further work to confirm these findings, their frequency and impact is needed. The breakup of summer arctic clouds is an established phenomena linked to the interaction of radiation with the stratocumulus cloud. However observations of the impact of secondary ice production in consumption of the liquid phase through vapour diffusion in these clouds is needed to understand whether this effect is significant.

The production of secondary ice through rime-splintering was the dominant secondary ice process observed in all the cases presented in this work. However other mechanisms that lead to enhancement in ice concentrations are possible, such as droplet shattering and the breakup of frail ice structures. Future research should aim to identify where these processes take place and their contribution to the glaciation of clouds.

Observations in natural clouds have found swift transitions between glaciated, liquid and mixed phase clouds. In all the papers discussed here transitions were also observed, with the measurements made at Jungfraujoch, Switzerland particularly notable for changes over differing temporal scales. Glaciation of clouds has been linked to changes in aerosol composition and concentration and at JFJ we found a possible link between fluorescent aerosol and ice concentrations. However more work is needed to quantify the influence of
aerosol types on ice concentrations and to assess the relative impact of dynamics, ice nuclei and surface processes at that particular measurement site.

8.3 Publications

The following is a list of publications associated with the work presented in this thesis.


CHAPTER NINE

REFERENCES


Foot, V.E. et al., 2008. Low-cost real-time multi-parameter bio-aerosol sensors. In J. C. Carrano & A. Zukauskas, eds. *International Society for Optics and Photonics*.


