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Observing aerosol impacts on Arctic clouds

Author:

Rebecca Murray-Watson

Supervisors:

Dr. Edward Gryspeerdt

Prof. Helen Brindley

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Abstract

Aerosol-cloud interactions represent a significant source of uncertainty in anthropogenic climate forcing, particularly in the Arctic where clouds critically influence the surface energy budget. The Arctic is undergoing rapid environmental transformations, and it is critical to understand how these changes affect cloud radiative properties to accurately project future regional climate shifts.

This thesis uses satellite data to develop constraints for aerosol-cloud interactions. Although satellites often struggle to obtain reliable measurements of Arctic clouds, this work shows that through careful filtering, the retrieval biases can be reduced. This work finds that the aerosol impacts on cloud liquid water path vary significantly across the region, predominantly controlled by lower tropospheric stability. As the stability is strongly influenced by the surface temperatures, this suggests a temperature-dependent aerosol effect. This could lead to a reduced cooling effect of clouds in a warmer, more polluted Arctic.

This work develops a novel method for creating pseudo-Lagrangian trajectories to study the temporal development of clouds as they move from the sea ice edge across the open ocean. By controlling for the initial conditions using reanalysis data, this work finds that the breakdown in the cloud field is strongly related to precipitation, which in turn is affected by the presence of aerosols through precipitation suppression.

Additionally, this thesis finds that the air mass history is strongly linked with the formation of ice along these trajectories. The predominance of mixed-phase clouds at temperatures as high as -13°C suggests that the availability of biological ice nucleating particles plays a central role. Overall, this thesis highlights the need for a

more detailed understanding of aerosol properties, sources and sinks in the region as a key target to improve our representation of Arctic clouds in climate models.

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Acronyms

AOD - Aerosol Optical Depth

AMSR-E - Advanced Microwave Scanning Radiometer for the Earth Observing System

ASCOS - Arctic Summer Cloud Ocean Study

CCN - Cloud Condensation Nuclei

CERES - Clouds and the Earth's Radiant Energy System

CF - Cloud Fraction

COMBLE - Cold-Air Outbreaks in the Marine Boundary Layer Experiment

CTP - Cloud Top Pressure

CTT - Cloud Top Temperature

DARDAR - raDAR/liDAR

DMS - Dimethylsulphide

EarthCARE - Earth Cloud Aerosol and Radiation Explorer

ECMWF - European Centre for Medium-Range Weather Forecasts

ECMWF-AUX - ECMWF Auxiliary Data

GCMs - General Circulation Models

HALO-(AC)³ - High Altitude Long range research aircraft - Arctic Amplification: Climate Relevant Atmospheric and Surface Processes, and Feedback Mechanism

IMS - Interactive Multisensor Snow and Ice Mapping System

INP - Ice Nucleating Particles

IPCC AR6 - Intergovernmental Panel on Climate Change Assessment Report 6

IWP - Ice Water Path

LTS - Lower Tropospheric Stability

LWP - Liquid Water Path

MCB - Marine Cloud Brightening

MCAO - Marine Cold Air Outbreak

MODIS - Moderate Resolution Imaging Spectroradiometer

MOSAIC - Multidisciplinary drifting Observatory for the Study of Arctic Climate

MSA - Methanesulfonic Acid

N_d - Cloud Droplet Number Concentration

NIR - Near-Infrared

RH - Relative Humidity

RWC - Rain Water Content

SHEBA - Surface Heat Budget of the Arctic Ocean

SPI - Subpixel Heterogeneity Index

SZA - Solar Zenith Angle

TOA - Top-of-Atmosphere

TSI - Time Since Ice

TTI - Time Towards Ice

WBF - Wegener-Bergeron-Findeisen

Statement of Originality

The following thesis is the product of my own research. All results or information taken from other work has been acknowledged and referenced appropriately.

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Chapter 1

Introduction

1.1 Aerosols, Clouds and Climate

Clouds play a central role in the Earth's climate system, regulating the surface energy budget by interacting with solar and terrestrial radiation. Clouds can warm the surface through absorption and re-emission of longwave thermal radiation, reducing the energy emitted to space relative to clear-sky conditions. Conversely, cloud scattering of incoming shortwave radiation serves to cool the planet by preventing surface heating. Overall, the albedo-enhancing effects of clouds are dominant and clouds exert a global net cooling effect (Loeb et al., 2009; Zelinka et al., 2017).

Cloud properties, such as their thermodynamic phase, altitude and abundance are crucial factors in determining their climate impact. These characteristics are closely dependent on cloud microphysical properties, such as droplet size and concentration (Lohmann et al., 2016). Cloud formation requires aerosols (liquid or solid atmospheric particles) to act as droplet nucleation sites, otherwise an unrealistically high atmospheric supersaturation must be achieved (Pruppacher and Klett, 1997). Aerosols may exert a significant influence on the cloud radiative forcing by modifying cloud droplet characteristics, which in turn change macroscale cloud properties.

These aerosol-cloud interactions are poorly constrained by observations, rendering them one of the largest uncertainties in understanding anthropogenic climate change (Forster et al., 2021).

The climate impact of aerosol-cloud interactions is split into two effects; the first is an instantaneous effect on cloud albedo and radiative properties (called RF_{aci} in Boucher et al. 2013), and the second is associated with the further responses of clouds to these aerosol perturbations, known as ‘adjustments’. A summary of these effects on both liquid and mixed-phase clouds is shown in Figure 1.1, and will be discussed further in the following sections.

1.1.1 RF_{aci}

Liquid Clouds This effect manifests primarily through the ability of aerosols to act as cloud condensation nuclei (CCN); increased aerosol loadings provides more nucleation sites, increasing the cloud droplet number concentration (N_d). If the cloud water content is unchanged, these droplets will be smaller as the water is distributed between more droplets (Twomey, 1974). The combined effect of enhanced N_d and smaller droplets, commonly known as the ‘Twomey effect’, increases the total surface area of the cloud droplets, enhancing the cloud albedo and amplifying the shortwave cooling effect (Twomey, 1977). For liquid clouds, this effect of reducing hydrometeor size but increasing N_d has been widely observed through satellite studies (e.g., Bréon et al., 2002) and in situ measurements (e.g., Feingold et al., 2003). However, despite the substantial quantities of research to date, models still produce a wide range of estimates for RF_{aci} due to the Twomey effect (Gryspeerd et al., 2020). While satellite data may provide observational constraints, there are significant challenges to overcome with their use (Quaas et al., 2020). For example,

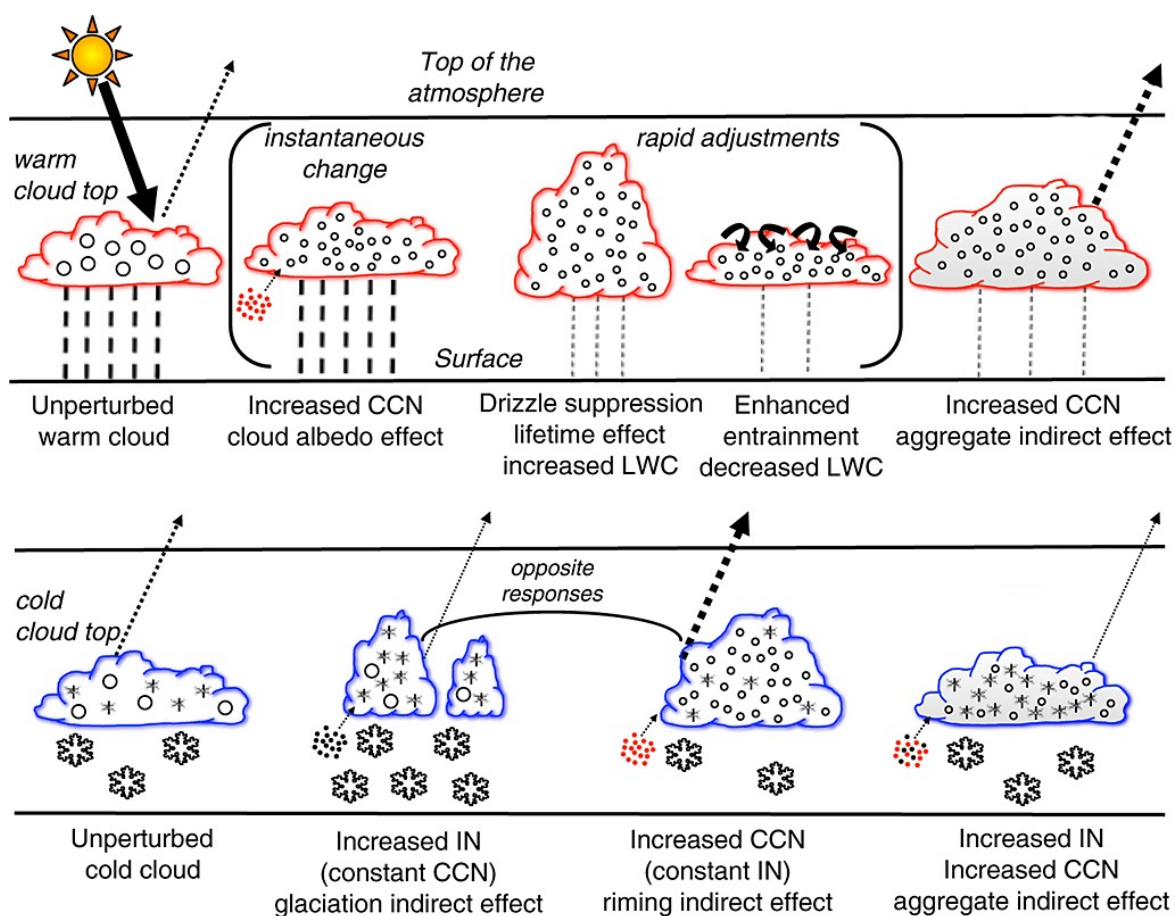


Figure 1.1: Graphical depiction of the different processes associated with aerosol-cloud interactions for liquid (red) mixed-phase (blue) clouds. The details of the mechanisms are described in the text. White circles represent cloud droplets and the red dots represent cloud condensation nuclei. The stars represent ice particles and the black dots are ice nuclei. The dashed arrows represent outgoing shortwave radiation, with the degree of emphasis indicative of magnitude. The dashed lines below the clouds indicates precipitation. Adapted from Christensen et al. 2014

satellites cannot measure the efficiency with which aerosols are activated as CCN at the cloud base, which is key to understanding the aerosol impact on clouds.

Mixed-Phase Clouds

Aerosols can also act as nucleation sites for ice crystals, and therefore can also regulate the properties of mixed-phase and ice clouds. The relationship is complicated by competing ice nucleation mechanisms and a lack of knowledge surrounding the

ability of anthropogenic aerosols to act as ice nucleating particles (INP) (Penner et al., 2018). Relative to liquid droplets, ice crystals are larger and have a smaller refractive index (Lohmann et al., 2016). Therefore, for a given cloud water content, clouds which have a mixture of ice and water have a lower optical thickness and a reduced albedo. Misrepresentation of the ratio of liquid to ice in mixed-phase clouds has been identified as a significant source of bias in the shortwave radiative fluxes in models (Tan and Storelvmo, 2019). The uncertainty surrounding the aerosol impacts on these clouds means that determining the net radiative effect is challenging, with many studies estimating the RF_{aci} from clouds omitting mixed-phase clouds entirely (Bellouin et al., 2020).

1.1.2 Adjustments

Liquid Clouds

Adjustments are largely realised through changes in the cloud liquid water path (LWP) or cloud fraction (CF), which are key properties when considering the cloud albedo (e.g., Engström et al., 2015). As an increase in aerosol loading creates smaller droplets, which coalesce less efficiently, aerosols can reduce the precipitation efficiency of clouds (Albrecht 1989; Rosenfeld 2000). This delay in cloud water loss may amount to a negative forcing through increasing the longevity of the cloud, thereby increasing the LWP and CF (Albrecht, 1989). However, smaller droplets also enhance the entrainment of air into the cloud; under dry conditions, these droplets undergo rapid evaporation. This evaporation generates further entrainment, thereby creating an evaporation-entrainment feedback and can decrease LWP and cloud cover (Ackerman et al., 2004). Buffering mechanisms in the system make the influence of aerosols on cloud properties difficult to deduce (Stevens and Feingold, 2009); for example, aerosol-induced instability may also deepen the cloud layer, creating

higher LWP clouds which in turn precipitate more, offsetting the LWP gains from the precipitation suppression mechanism.

The competing effects of precipitation suppression and evaporation-entrainment make isolating the aerosol effects on CF and LWP challenging. Identifying a causal relationship is further confounded by local meteorology; in particular, free tropospheric relative humidity (RH) and lower tropospheric stability (LTS), which is a measure of the thermodynamic stability of the atmosphere (Klein and Hartmann, 1993), are found to be important when considering the aerosol-LWP relationship (e.g., Chen et al., 2014). Many studies find the sign of the LWP response is contingent on meteorological conditions (Han et al., 2002; Ackerman et al., 2004; Lebsock et al., 2008; Chen et al., 2014; Michibata et al., 2016; Gryspeerdt et al., 2019a), with decreases typically seen in unstable environments with low cloud-top humidity due to the entrainment of dry air. However, increases were also observed under high cloud-top RH conditions. Furthermore, in precipitating clouds, an increase in LWP was observed, which is consistent with precipitation suppression (e.g., Lebsock et al., 2008; Sorooshian et al., 2009; Chen et al., 2014).

Natural experiments, in which the aerosol source is independent of meteorology, allow for a direct investigation of aerosol-cloud interactions (Christensen et al., 2022). These studies, which include analysis of ship and volcano tracks, have generally found a weakly negative LWP response on average, which offsets the negative forcing of RF_{aci} (e.g., Toll et al., 2017; Malavelle et al., 2017; Toll et al., 2019). These bidirectional responses observed through satellite imagery are in contrast to the increase in LWP often simulated by climate models due to the parameterisations of precipitation suppression (e.g., Toll et al., 2017). Careful interpretation of these studies is required, as they are based on a ‘snapshot’ analysis of satellite imagery, in which instantaneous correlations between different variables is calculated based on

many independent satellite images. As a result, the time-dependent nature of cloud adjustments is not considered. Recent work proposes that evaluating the temporal evolution of aerosol-cloud interactions is key to achieving a process-level understanding (Gryspeerdt et al., 2021).

Mixed-Phase Clouds

The scientific understanding of adjustments in mixed-phase clouds is less advanced than those in liquid clouds due to the complexity of aerosol-ice interactions and limited observational constraints. An increase in anthropogenic INP may induce a phase transformation in mixed-phase clouds; as the saturation vapour pressure of ice is lower than that of liquid water, ice crystals grow at the expense of water droplets. This mechanism, known as the Wegener-Bergeron-Findeisen (WBF) process (Wegener, 1911; Bergeron, 1935; Findeisen, 1938), makes the cloud thermodynamically unstable and may rapidly convert cloud liquid water to ice crystals and result in total cloud glaciation (Lohmann et al., 2016; Korolev et al., 2017). Enhanced precipitation of these ice crystals results in lower cloud cover and an increase in solar radiation reaching the surface, contributing to a positive forcing (Jiang et al., 2000; Lohmann, 2002). There is some observational evidence for the glaciation of mixed-phase clouds by anthropogenic aerosols (e.g., Christensen et al., 2014; Coopman et al., 2018b), but the mechanism is not well understood. Vergara-Temprado et al. 2018 showed that the inability to realistically simulate INP concentrations was a significant contributor to the discrepancies between modelled and observed mixed-phase cloud cover over the Southern Ocean, with associated uncertainties in their impact on climate. An increase in CCN may counteract the glaciation effect in mixed-phase clouds by reducing droplet size and making riming less efficient, thereby reducing water loss through precipitation (Borys et al., 2003). However, there is little observational evidence for this effect and it may have a relatively weak

influence on cloud lifetime (Lohmann, 2004, 2017). A ‘thermodynamic’ effect has also been proposed, in which smaller droplets freeze less efficiently (Lohmann and Feichter, 2005). Lance et al. 2011 suggested that ice formation was suppressed in polluted Arctic clouds due to a shift to smaller droplets, although they failed to quantify the effect due to uncertainties surrounding the ice nucleation mechanism.

The overall effect of aerosols mixed-phase cloud lifetimes is challenging to determine, in large part due to a lack of observations of INP, ice formation processes and confounding meteorological variables (Korolev et al., 2017; Bellouin et al., 2020). Estimating the magnitude of the associated anthropogenic forcing is further hindered by these uncertainties and a lack of knowledge of future INP states (Murray et al., 2021). Overall, best estimates suggest that it is a positive forcing (net warming; Bellouin et al. 2020), although global observational constraints are needed to reduce inter-model variation.

The latest Intergovernmental Panel on Climate Change (IPCC AR6; Forster et al. 2021) report determined that the total radiative forcing generated by aerosol-cloud interactions (ERF_{aci}) to be -0.84 W m^{-2} (-1.45 to -0.25 W m^{-2} at a 90% confidence interval, change from 1750 to 2019). The significant uncertainty range is reflected in the ‘medium’ level of confidence assigned to this estimate ERF_{aci} .

1.1.3 Arctic Clouds and Environment

Arctic Energy Budget

The Arctic is undergoing rapid change, warming at a rate between twice and four times as fast as the rest of the globe (Serreze and Barry, 2011; Rantanen et al., 2022). This heating has considerably altered the Arctic environment, most notice-

able in the swift decline in sea ice extent. Arctic sea ice cover has been decreasing at about 12% per decade since records began, with eight of the ten lowest September Arctic sea ice extents on record having occurred since 2010 (National Snow and Ice Data Center, 2023). The region is projected to be entirely devoid of summer sea ice some time between 2030 and 2050 (Overland et al., 2019; Guarino et al., 2020). Such a profound shift will have considerable economic, environmental and cultural impacts in the region (e.g., Durkalec et al., 2015). Additionally, sea ice loss may lead to changes in midlatitude weather patterns (Coumou et al., 2018; Smith et al., 2022).

Anthropogenic greenhouse gas emissions are the primary cause of increased temperatures in the Arctic. However, various feedback mechanisms are responsible for the accelerated heating of the Arctic, a phenomenon known as Arctic Amplification. Of these feedbacks, the surface albedo feedback is often cited as a significant contributor to enhanced Arctic temperatures; as the cryosphere warms, highly reflective surfaces such as sea ice (albedo of up to 1) are lost. Lower albedo surfaces, such as the ocean (albedo of around 0.1), are exposed instead. These dark surfaces absorb substantially more incoming shortwave solar radiation than the original, high albedo surfaces, which contributes to further heating (Curry et al., 1995; Screen and Simmonds, 2010; Kashiwase et al., 2017). This cycle becomes a positive feedback loop, promoting further surface warming and sea ice loss. However, Pithan and Mauritsen (2014) found that while the surface-albedo feedback was important, temperature and lapse rate feedbacks were more significant contributors to Arctic Amplification.

Arctic clouds are also involved in feedback pathways, although their exact influence on Arctic Amplification is uncertain. One mechanism of influence is linked to sea ice loss; as more open ocean is exposed, there can be an increase in transport of heat and moisture from the surface to the atmosphere. This increase in fluxes can

generate more low-level cloud cover (Schweiger et al., 2008; Kay and Gettelman, 2009; Morrison et al., 2019). The net radiative effect of these clouds depends on the season. During much of the year, and especially during polar night when much of the Arctic experiences little incoming solar radiation, the longwave radiative effect dominates and these clouds warm the surface; relative to clear sky conditions, the net longwave forcing can reach $40\text{--}50\text{ W m}^{-2}$ (Curry et al., 1996; Intrieri et al., 2002; Shupe and Intrieri, 2004). This enhances the positive sea ice-albedo feedback. An increase in cloudiness during the spring has been linked to decreases in sea ice extent (Kapsch et al., 2013). Similarly, autumnal cloud coverage is strongly linked to sea ice refreezing, with increases in coverage linked to lower sea ice extents (Kay and Gettelman, 2009).

Conversely, during the polar day, with sufficient incoming solar radiation, clouds can effectively buffer the loss of the highly reflective sea ice by acting as a high-albedo surface over the ocean and creating a negative feedback (e.g., He et al., 2019). However, satellite-based studies have shown that due to weak ocean-atmosphere coupling, there is little increase in surface fluxes during the summer months following sea ice loss, so cloud cover does not strongly increase despite the exposed ocean surface (Kay and Gettelman, 2009; Morrison et al., 2019). Although there is no summertime cloud-sea ice feedback, summertime clouds can play an important role in regulating further sea ice loss. Increases in the amount of solar radiation absorbed by the surface, which is controlled both by the cloud coverage and properties such as the cloud water path is linked with decreases in sea ice coverage later in the year (Choi et al., 2014; Huang et al., 2017, 2019). In particular, Kay and Gettelman (2009) found the increased surface warming caused by reductions in summertime cloud coverage enhanced the sea ice-albedo feedback and contributed to the then-record sea ice loss in 2007 (which remains the joint third-lowest sea ice extent on record; National Snow and Ice Data Center 2023).

The second feedback mechanism is associated with cloud phase and optical depth. As the Arctic near-surface temperatures increase, ice formation is suppressed in the boundary layer, and mixed-phase clouds shift to a state with higher liquid fractions (Mitchell et al., 1989). As liquid droplets are more reflective to shortwave radiation than ice for a given mass of water, these clouds have become optically thicker and have a higher albedo and therefore induce a net cooling effect. Additionally, the adiabatic cloud water amount is projected to increase with warming, further increasing the cloud optical depth (e.g., Ceppi et al., 2016). This constitutes a negative feedback over low albedo surfaces. However, the strength of the feedback with warming is uncertain, with a strong dependence on how models partition liquid and ice phases in mixed-phase clouds (Storelvmo et al., 2015). Part of this uncertainty is due to the way in which mixed-phase clouds are currently represented in climate models, with climate models often underestimating the supercooled liquid and overestimating the ice fraction in present-day mixed-phase clouds (Tan et al., 2016; Bodas-Salcedo et al., 2016). This overestimation of present-day ice fraction implies that the magnitude of the negative feedback observed in climate models may be too strong, and therefore underestimate climate warming. Zelinka et al. (2020) suggest that one cause of higher climate sensitivity (the response of temperature due to a doubling of CO₂ concentrations relative to pre-industrial levels) seen in the sixth Coupled Model Intercomparison Project (CMIP6) relative to CMIP5 is due to the improved representation of mixed-phase clouds, in particular their phase partitioning. This effect is likely to be regulated by present and future concentrations of INPs, which are also poorly constrained (Murray et al., 2021).

Overall, the role of clouds is one of the most poorly understood aspects of Arctic amplification (Goosse et al., 2018), with some studies finding a strong positive enhancement of warming (Vavrus, 2004), and others finding little effect (e.g., Screen

and Simmonds, 2010). Using models from CMIP5, Pithan et al. (2014) found a small positive contribution, although the models disagreed on the sign and magnitude of the feedback. Part of the reason for this uncertainty is due to the difficulty in representing clouds in climate models; Lenaerts et al. 2017 found a large variation in cloud characteristics between models used in CMIP5, especially in representing cloud liquid and ice water paths (LWP and IWP). Due to their importance in the surface energy budget, it is imperative to realistically represent cloud phase, coverage and properties. The inability of models to capture cloud properties leads to difficulties in representing the cloud-sea ice relationship (e.g., Huang et al., 2019), and contributes to inter-model spread in sea ice projections (Luo et al., 2023). In particular, a key difficulty in resolving these issues is cited as the uncertainty surrounding the representation of aerosol-cloud interactions in climate models, an issue that exists with modelling clouds globally. Use of remote sensing data has been cited as essential to addressing these uncertainties and improving cloud parameterisations (Kay et al., 2016; Lenaerts et al., 2017).

Characteristics of Arctic Clouds

In situ and satellite studies reveal a distinct seasonal cycle of maximum cloudiness in late summer or early autumn (often exceeding 90%) and a minimum in spring (e.g., Shupe, 2011; Boeke and Taylor, 2016). In particular, low-level (below 3 km) liquid and mixed-phase are ubiquitous (Shupe 2011; Bennartz et al. 2013; Mioche et al. 2015; Kay et al. 2016; Figure 1.2) and play a key role in the surface energy budget (Shupe and Intrieri, 2004). Low clouds often form when warm air masses from lower latitudes are advected over cold Arctic ocean (Herman and Goody, 1976; Klein and Hartmann, 1993; Pithan et al., 2018). These air masses radiatively cool to saturation point, allowing low-level stratiform clouds to form. Surface-atmosphere coupling can also be important for cloud formation (Kay and Gettelman, 2009; Kay et al., 2016); thermodynamically unstable boundary layers facilitate the vertical transport

of moisture and latent heat, creating conditions suitable for cloud formation. This mechanism shows some seasonal dependence; during the summer melt season, the ocean remains near melting temperature while the overlying air warms (Persson, 2012). This stabilises the atmosphere, impeding ocean-atmosphere coupling and summertime cloud development (Morrison et al., 2019). During autumn, the ocean warms while the atmosphere cools, lowering static stability and enhancing upward heat and moisture fluxes and promoting cloud formation (Kay and Gettelman, 2009; Boisvert et al., 2015).

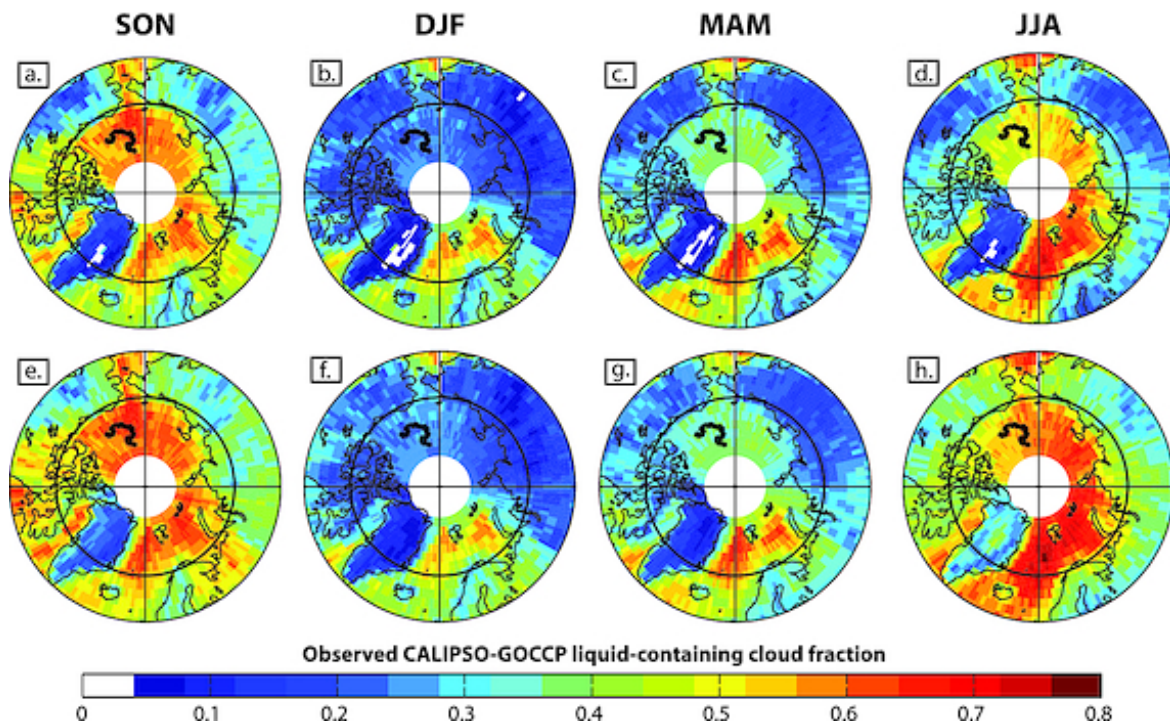


Figure 1.2: Seasonal cycle of liquid-containing cloud fraction from CALIPSO-GOCCP observations for 2006-2011. Figures (a) to (d) are for low level clouds (below 3.36 km) and Figures (e) to (h) are for all altitudes. From Cesana et al. 2012.

The prevalence and persistence of mixed-phase clouds are important characteristics of the Arctic climate system. These clouds comprise both supercooled liquid droplets and ice crystals, with in situ measurements showing an increase in the liquid fraction with temperature, although substantial amounts of liquid may be present at temperatures significantly below freezing (Shupe et al., 2006; Cesana et al., 2012).

Despite the inherent thermodynamic instability of the ice-liquid mixture and mass loss due to ice precipitation, these clouds can exhibit surprising longevity, lasting up to several days (Shupe et al., 2006; Shupe, 2011). This persistence is in part due to a self-maintaining feedback driven by longwave radiative cooling of the cloud top, where most of the supercooled liquid droplets reside (Morrison et al. 2012; Figure 1.3). This cooling promotes turbulence in the boundary layer, generating updrafts which become supersaturated with respect to ice and liquid water, maintaining the cloud.

The advection of moist air masses from lower latitudes into the Arctic often creates conditions in which the specific humidity increases with height above the cloud top (Curry et al., 1996; Tjernström et al., 2004; Sedlar et al., 2012). This feature is in stark contrast to lower-latitude stratocumulus clouds, which often sit beneath a layer of dry air (e.g., Klein and Hartmann, 1993). Therefore, while entrainment may be expected to evaporate subtropical clouds, Arctic clouds may instead increase their total water content. This cloud-top moisture source is particularly important in sustaining the cloud in cases where the cloud layer is decoupled from the surface moisture sources (Solomon et al., 2011; Sedlar et al., 2012; Morrison et al., 2012).

As discussed in Section 1.1.2, the phase partition of Arctic clouds is crucially important to their climate impact due to the different radiative properties of ice particles and liquid water droplets (Sun and Shine, 1994). Clouds with higher supercooled liquid fractions are both more opaque to thermal radiation and reflect more short-wave radiation than ice crystals (Sun and Shine, 1994; Hogan et al., 2003). The relative abundance of ice particles and liquid droplets can also influence other cloud properties, such as cloud lifetime, which in turn affects the net radiative effect.

However, many climate models underestimate the supercooled liquid fraction of

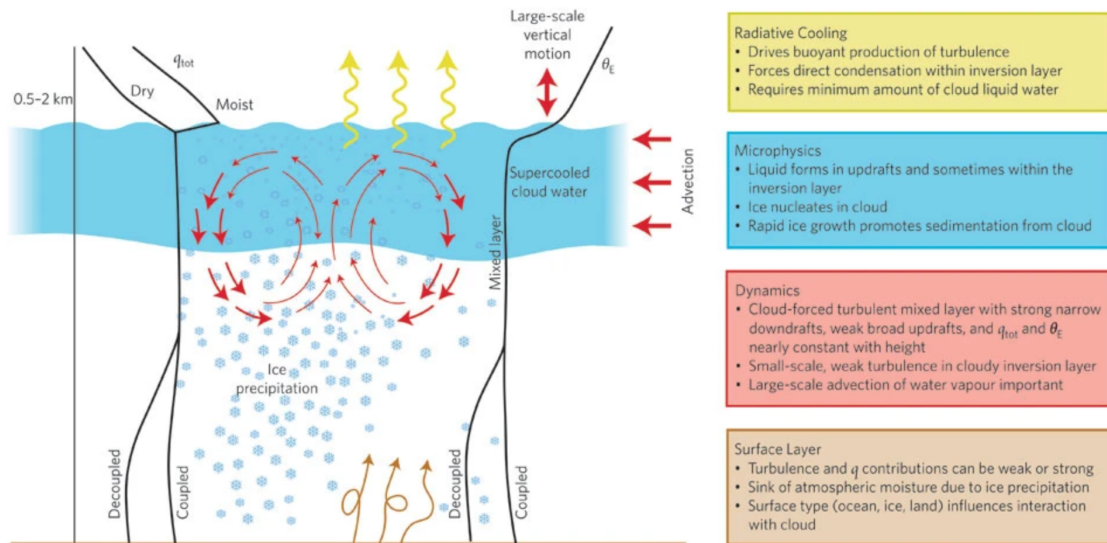


Figure 1.3: Seasonal cycle of liquid-containing cloud fraction from CALIPSO-GOCCP observations for 2006-2011. Figures (a) to (d) are for low level clouds (below 3.36 km) and Figures (e) to (h) are for all altitudes. From Cesana et al. 2012.

mixed-phase clouds, leading to significant uncertainties in net radiative effect of these clouds (Cesana et al., 2012, 2015; Tan and Storelvmo, 2019). This in part arises from a lack of observations; in situ measurements and field campaigns are naturally restricted in space and time, so while they have provided invaluable insights into mixed-phase cloud processes, significant uncertainties remain (Korolev et al., 2017). While remote sensing, in particular active satellites, allow for the collection of long-term statistics over the whole region, obtaining a mechanistic understanding of the dominant controls on cloud formation and lifetime is still challenging and certain cloud characteristics, such as droplet concentration, cannot be retrieved directly. Additionally, the coarse resolution of general circulation models (GCMs) means they struggle to simulate processes such as the WBF mechanism due to the microphysical scales on which they occur (e.g., Pithan et al., 2014). Several studies showed that models which included more sophisticated microphysics parameterisations generally performed better at simulating the cloud phase partition than those using a simple temperature-dependent scheme (Cesana et al., 2012; McCoy et al., 2016; Cesana et al., 2022). When comparing CMIP5 models, Taylor et al. (2019) found that dif-

ferences in microphysics schemes, in particular those related to ice formation and phase partition, were largely responsible for the inter-model discrepancies in Arctic cloud cover. Inaccuracies in simulating the supercooled liquid fraction can affect projections of climate sensitivity and Arctic amplification (Tan et al., 2016; Tan and Storelvmo, 2019), underscoring the need for both observational constraints and better representations of cloud phase in models.

Arctic aerosols

As aerosols can strongly influence the radiative properties of clouds, understanding their sources and sinks in the Arctic climate is of great importance to improving modelling efforts. However, there is significant uncertainty surrounding the current and future state of Arctic aerosols (Schmale et al., 2021). The Arctic typically has low CCN concentrations relative to other regions, with values reaching as low as 1 cm^{-3} in the central ice pack (Mauritsen et al., 2011). INP, which are naturally less abundant than CCN, are also particularly scarce in the Arctic (Bigg, 1996; Wex et al., 2019). The aerosol burden exhibits a strong seasonal cycle, typically reaching a peak during boreal winter and a minimum during summer months (Tunved et al., 2013; Schmale et al., 2022a). The winter maximum, often called the ‘Arctic haze’, is due to substantial long-range transport of anthropogenic pollution from lower latitudes and a lack of wet removal processes (Shaw, 1995; Shen et al., 2017; Willis et al., 2018; Boyer et al., 2023). In contrast, natural aerosols are dominant during the summer, particularly from biogenic sources such as phytoplankton activity near the sea ice edge (Willis et al., 2018). These phytoplankton blooms produce dimethylsulphide (DMS) and methanesulfonic acid (MSA), which are key drivers of particle formation (Chang et al., 2011; Willis et al., 2017). In particular, INP are more abundant in spring and summer due to the greater presence of snow- and ice-free marine and terrestrial sources (Wex et al., 2019).

While in situ and field studies have collected important data about aerosol chemistry and morphology, characteristics which dictate their ability to act as CCN or INP, these are sparse and of limited geographic extent. As the Arctic is a vast, heterogeneous area with a large variety of different aerosol formation processes, it is uncertain how representative these measurements are of the whole region (e.g., Freud et al., 2017). Passive satellite instruments, such as the Moderate Resolution Imaging Spectroradiometer (MODIS; Section 2.1) usually fail to obtain valid retrievals of Arctic aerosol loadings due to their reliance on both incoming solar radiation and dark surface targets (Levy et al., 2009). Active sensors can instead provide information about aerosol concentrations and vertical extent, which is important when considering their interactions with clouds. However, they have very limited footprints and therefore restricted coverage.

These knowledge deficits mean that models struggle to accurately represent Arctic aerosol processes (e.g., Eckhardt et al., 2015; Schmale et al., 2021). Both natural and anthropogenic aerosol concentrations are projected to change in the future. Sea ice retreat and ocean warming may lead to an increase in Arctic biogenic activity, which may in turn increase summertime CCN concentrations (Willis et al., 2017). Increased exposure of open water may also increase the flux of sea salt aerosols to the boundary layer (Struthers et al., 2011). Some regions are projected to undergo significant development in the coming decades due an increase in the exploitation of natural resources in the Arctic (Schmale et al., 2018). This increase in industrialisation, in conjunction with increased shipping due to the opening of new trans-Arctic trade routes (Peters et al., 2011), may increase the anthropogenic aerosol load. However, future changes in precipitation may increase the wet scavenging of aerosols and therefore reduce transport from long-range sources (McCrystall et al., 2021). The total effects on Arctic clouds and climate remain unclear due to seasonal

variability, dependency on economic development and uncertainties about the current baseline state of aerosols in the region.

Aerosol-cloud interactions in the Arctic

Despite the importance of both clouds and aerosols in the high-latitude climate system, the considerable challenges associated with their study means that there are still substantial uncertainties surrounding their interactions. However, a number of in situ studies or field campaigns have made some progress in addressing these knowledge gaps. When using ground-based in situ instruments based at Barrow, Alaska, Garrett et al. 2004 found that Arctic clouds were more sensitive to aerosols than lower-latitude clouds, with particularly strong changes in cloud microphysical properties during Arctic haze events. Koike et al. 2019 also found that Arctic cloud properties were strongly sensitive to aerosol properties, but found a similar response across all seasons. Garrett and Zhao 2006 found that these Arctic haze events increased the emissivity of greybody clouds (LWP below 40 g m^{-2}) as the smaller, more numerous droplets interacted more strongly with longwave radiation. The emissivity of thicker clouds was insensitive to pollution as they already radiated as blackbodies. Estimates of the longwave forcing associated with this increase in emissivity in these optically thin clouds vary in magnitude ($2\text{-}5.2 \text{ W m}^{-2}$; Garrett et al. 2004; Garrett and Zhao 2006; Lubin and Vogelmann 2006), but indicate that these Arctic haze events may amplify surface heating. However, Zhao and Garrett 2015 found aerosol-cloud interactions had a net cooling effect on the surface at Barrow from May through September, with up to 70% of the changes in radiative forcing measured were caused by the Twomey effect, with the rest due to LWP increases.

Arctic haze events represent periods of significant aerosol loading; however, some high latitudes regions have very low aerosol concentrations, which hinders cloud

formation. Using measurements of the central Arctic Ocean taken during the Arctic Summer Cloud Ocean Study (ASCOS), Mauritsen et al. 2011 found that even moderate increases in CCN availability for this ‘tenuous cloud regime’, which regularly has CCN concentrations below 10 cm^{-3} , can substantially increase the cloud long-wave effects and augment surface warming. Furthermore, these low aerosol levels may be maintained by a positive feedback loop due to the efficient precipitation of larger cloud droplets, which in turn removes aerosols through wet deposition. This aerosol-limited cloud regime has been observed by other studies in the high Arctic (Engelmann et al., 2021; Guy et al., 2021; Pasquier et al., 2022) and has been linked to the rapid dissipation of mixed-phase clouds (Sterzinger et al., 2022).

While in situ, field and aircraft campaigns have been invaluable in the study of Arctic aerosol-cloud interactions, significant drawbacks include their limited spatial coverage and potentially restricted timespan. Satellites are powerful tools to overcome these issues of low spatial and temporal coverage (Kay et al., 2016). While satellites have been widely used to study Arctic clouds (e.g., Cesana et al., 2012) and their response to sea ice retreat (e.g., Kay and Gettelman, 2009), there has been relatively little work using these data to examine aerosol-cloud interactions. This is in part due to the aforementioned difficulties in retrieving reliable aerosol data in the Arctic. In terms of cloud retrievals, passive sensors rely on contrast between their targets and the underlying surface, and therefore have difficulty distinguishing clouds from cold, bright surfaces such as snow and sea ice. Additionally, passive sensors struggle to identify mixed-phase clouds, which if erroneously classified as a liquid cloud may bias cloud property retrievals (e.g., Khanal and Wang, 2018). Issues using passive instruments to study cloud properties are discussed further in Chapter 3.

The difficulties with detecting aerosol concentrations have previously been circumvented by using passive tracers from a chemical transport model in lieu of aerosol

retrievals and co-locating these data with satellite-derived cloud properties (Tietze et al., 2011). These passive tracers act as proxies for CCN concentrations in pollution plumes, such as those generated by anthropogenic industrial sources. Coopman et al. 2016 investigated the impact of anthropogenic aerosols transported from mid-latitudes on Arctic clouds using a carbon monoxide (CO) tracer simulated by the FLEXPART model. Coopman et al. 2016 found that Arctic cloud optical depth and droplet size are highly sensitive to aerosol, with meteorological conditions exerting a strong control on the response. However, the LWP was only weakly sensitive to aerosol perturbations. Using a similar technique, Coopman et al. 2018a compared the effects of anthropogenic pollution with aerosols generated by biomass burning. They found that clouds were more susceptible to industrial pollution than biomass burning aerosols, but could not fully explain why. A weak LWP response was also observed, although other cloud properties were strongly modified by aerosols. Using industrial emissions as a natural experiment, Maahn et al. 2021 found increases in shortwave cooling due to the Twomey effect in liquid and mixed-phase clouds in summertime Alaska, but could not link these aerosols to changes in LWP or cloud fraction.

Coopman et al. 2018b, again using a passive CO tracer, found polluted clouds with smaller droplets freeze at lower temperatures due to the larger energy barrier for droplet glaciation. In spite of this, they observed an overall decrease in the energy barrier with higher levels of pollution, suggesting that some other unidentified property of the plumes is conducive to ice nucleation. Zamora et al. 2018 also found evidence of combustion aerosols triggering cloud glaciation during polar night by using active satellite sensors and the FLEXPART model to estimate black carbon aerosol loading. Overall, they found that meteorological variables were the dominant control on cloud properties, with variations in temperature and relative humidity explaining more than half of the variability in cloud fraction. However,

they also observed a decrease in low level cloud fraction, increased amounts of ice-containing clouds and increases in precipitation with increased aerosol concentrations. Although this points to combustion aerosols triggering ice nucleation, the exact microphysical mechanisms could not be identified from these data alone.

While implementing modelled aerosols or tracers avoids dealing with the difficulties with satellite-retrieved aerosol measurements, they are associated with their own uncertainties. A lack of knowledge about aerosol processes in the Arctic means models struggle to accurately simulate aerosol characteristics or concentrations, particularly in remote regions with few observational constraints (Schmale et al., 2021). This could serve as a potential source of error in the above results, although consideration of the relative changes in cloud and aerosol properties mitigates this (Coopman et al., 2016, 2018a). These works also all focus on the effects of long-range pollution on Arctic clouds. While this is important, particularly during boreal winter, the response to local aerosol sources must also be constrained.

1.1.4 Thesis Outline

It is evident that there is a considerable lack of knowledge surrounding aerosol-cloud interactions in the Arctic. This thesis aims to address this deficit by using state-of-the-art satellite data to probe the relationship between aerosols and cloud properties. These data are combined with reanalysis products to investigate the effects of meteorology on the aerosol-cloud relationship. This work also considers the temporal development of Arctic clouds using satellite data, a topic which is poorly understood but is crucial to improving model parameterisations of cloud processes.

The thesis is structured as follows:

Chapter 2

This chapter provides an overview of the data products used in the remainder of this thesis. In particular, this chapter discusses the strengths and potential weaknesses of the data, and why they are suitable for addressing the research questions in this work.

Chapter 3

The LWP derived from MODIS is compared against passive microwave retrievals to identify biases in the MODIS data. This chapter discusses sampling strategies to select for cases in which the MODIS algorithm can reliably retrieve cloud properties, justifying its use in the remainder of this thesis.

Chapter 4

This chapter considers the sensitivity of cloud LWP to aerosols, which is one of the most uncertain aspects of aerosol-cloud interactions. This is done by using N_d as a mediating variable, as aerosols act through cloud droplets to change other cloud properties. The influences on the N_d -LWP relationship are investigated, and how the radiative forcing from this relationship may change in a warmer, more polluted Arctic, is considered.

Chapter 5

A novel method to consider the temporal development of clouds moving away from the ice edge is developed. This chapter examines how the properties of liquid-dominated clouds evolve and are influenced by the initial conditions experienced along their trajectories.

Chapter 6

This chapter expands on the previous chapter's work by considering the evolution of mixed-phase clouds, in particular the factors affecting the formation of ice in these clouds. The trajectory analysis developed in Chapter 5 is extended to consider air mass history and the role this may have in the availability of INP.

Chapter 7

This chapter summarises the key findings and discusses potential future research avenues to address the limitations encountered in this thesis.

Chapter 2

Data

2.1 MODIS

Data from the Moderate Resolution Imaging Spectroradiometer (MODIS) Level 2 Collection 6.1 data product (MYD06_L2; Platnick et al., 2017) underpin the majority of this work. There are two satellites flying the MODIS sensor; Terra and Aqua, sun-synchronous satellites which have overpass times of 10:30 A.M and 1:30 P.M local solar time, respectively. Their polar-orbiting path means that most locations at lower latitudes are only observed twice a day, which limits the study of cloud temporal development. However, the Arctic is observed much more frequently; some locations may be observed more than 15 times in a 24 hour period. With 36 bands and providing data at 1 km resolution, MODIS provides state-of-the-art data which has supported a significant proportion of recent work in cloud physics.

MODIS cloud property retrievals start with the MODIS 1 km resolution cloud mask (Ackerman et al., 2010). The cloud mask differentiates between clear-sky pixels and cloudy using a decision tree based on 22 of MODIS's visible, near-infrared and infrared channels, with factors such as surface type determining which channels are used. Each pixel is assigned one of four values; confidently cloudy, probably cloudy,

probably clear and confidently clear. The cloud fraction is then determined as the ratio of the confidently and probably cloudy pixels to the confidently and probably clear pixels for the 25 pixels making up the 5 km product. The MODIS cloud detection algorithm can struggle to discriminate between clouds and snow- or ice-covered surfaces, in particular due to their similarity in the visible spectrum, leading to significant biases relative to active satellites (Chan and Comiso, 2013). However, due to greater thermal and visible contrast between clouds and the surface, MODIS performed better over ocean, with only 3.7% disagreement in cloud detection between MODIS and active sensors. Given these constraints, this thesis will concentrate on analysing cloud cover over oceanic regions during the polar day. The MODIS data used in this work spans 2008 to 2015 and were regridded from their native resolution (either 1 km by 1 km or 5 km by 5 km) to a 25 km by 25 km polar stereographic grid. This grid is used for nearly all of the analysis presented in this thesis.

2.1.1 Cloud Property Data

2.1.2 r_e and τ_c

Knowledge of cloud microphysical properties, namely N_d and LWP, is essential for studies of aerosol-cloud interactions. These are not directly retrieved by MODIS, but can be derived (using assumptions described below) from retrievals of cloud effective radius (r_e) and cloud optical depth (τ_c). These cloud properties can be retrieved using radiometric measurements in the visible and near-infrared (NIR) bands (Nakajima and King, 1990), shown in Figure 2.1. NIR bands 1.6 μm , 2.1 μm and 3.7 μm , which have different penetration depths into the cloud (Platnick, 2000), can be used, but the 2.1 μm data form the standard MODIS r_e product which is used in this thesis. While both τ_c and r_e are sensitive to the NIR bands, only τ_c has strong variability with the visible wavelengths (over-ocean retrievals use 0.86 μm).

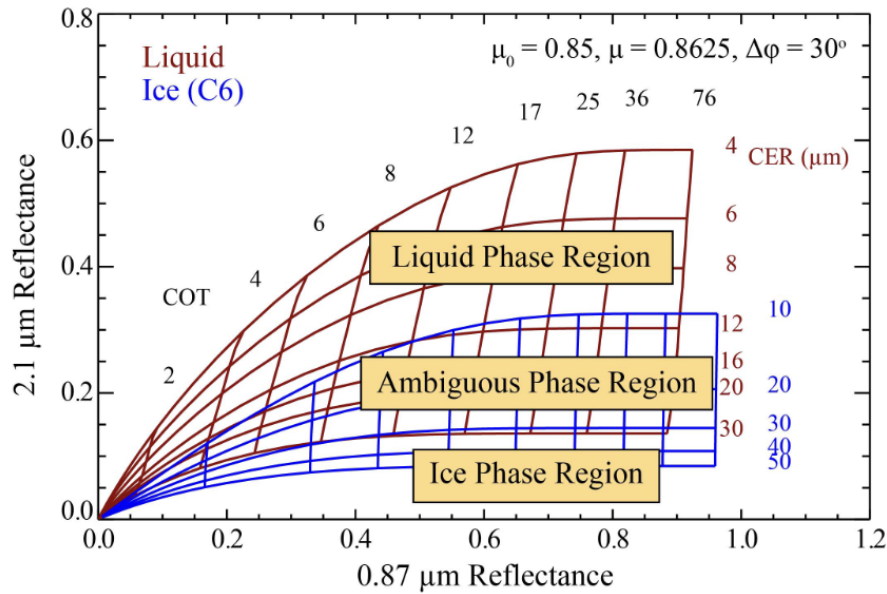


Figure 2.1: Nakajima and King-style diagram for ice and liquid clouds at 2.1 and 0.87 μm . Taken from Platnick et al. 2017.

One of the key assumptions underpinning the r_e and τ_c retrieval is the plane parallel approximation (Nakajima and King, 1990). This assumption stipulates that clouds are horizontally homogenous at both sub- and inter-pixel scales, such that there is no horizontal photon transport and pixels are not affected by their neighbours (the independent pixel approximation). This assumption simplifies the radiative transfer to a 1-D calculation. However, this assumption can break down in real-world scenarios, as clouds can vary significantly in their horizontal structure, particularly in broken cloud fields. This leads to 3D radiative effects, such as the transfer of photons between pixels, leading to retrieval biases. The plane parallel approximation is more accurate in high-coverage, stratiform clouds (Wood and Hartmann, 2006; Di Girolamo et al., 2010) and selecting for regions of high cloud fraction from the MODIS data ($>90\%$) has previously been used as a strategy to select for homogeneous scenes (Painemal and Zuidema, 2011); this is discussed further in Chapter 3. The MODIS cloud subpixel heterogeneity index (SPI), which is the ratio of the standard deviation to the mean of the reflectance in the 0.86 μm band of the 250 m

pixels which comprise each 1 km pixel, is a measure of the sub-pixel heterogeneity produced by the MODIS data product team. Larger values of SPI (>0.3) are associated with heterogeneous cloud regimes, such as broken cumulus fields (Zhang and Platnick, 2011).

One issue with the plane-parallel approximation that is particularly salient in the Arctic is its angular dependence, as it breaks down in cases with high solar zenith angle (SZA), which are common at high latitudes. At large SZAs, the 3D radiative effects associated with cloud heterogeneity, such as edge illumination or shadowing effects, become more prominent, and therefore lead to retrieval biases. Loeb et al. (1997) found that the break down of the plane parallel assumption at high SZA leads to significant increases in τ_c with SZA. Grosvenor and Wood (2014) found that below SZA of 65° , neither τ_c nor r_e showed significant variation with SZA. However, at higher angles, the biases rapidly grew with a positive bias of 70% in τ_c and negative bias of 5% for r_e . These lead to errors in N_d of up to 70%.

Another issue arises in cases of thin cloud layers, as the surface can contribute to the visible wavelength reflectance and therefore the microphysical retrievals (Platnick et al., 2017). This may be particularly important in cases of high-albedo surfaces, such as snow or ice, when the surface contribution may be strong enough that low optical depth clouds are not detected (e.g., King et al., 2004; Chan and Comiso, 2013). However, this contribution is less significant in cases of less reflective surfaces, such as the open ocean surface (Rosenfeld et al., 2004). Although clouds over snow- and ice-covered surfaces are important for the surface energy balance and can play a role in melting processes (e.g., Kay and Gettelman, 2009; Bennartz et al., 2013), the retrievals of cloud microphysical properties over these surfaces are highly uncertain. Hence, clouds over the ice-free ocean will be the focus of the work presented here. Additionally, at low τ_c and r_e , the retrievals are degenerate (Figure

2.1), and therefore are not independent retrievals. Cases with $\tau_c > 4$ and $r_e > 4 \mu\text{m}$ are considered more reliable (Quaas et al., 2006).

When using r_e to derive other cloud properties using the adiabatic assumption (discussed below), it is assumed that the r_e increases monotonically from the cloud base to the cloud top. The r_e retrievals are only sensitive to the upper portion of the cloud as even the most penetrating wavelength, $1.6 \mu\text{m}$, only reaches a few optical depths into the cloud. However, processes such as entrainment can reduce the r_e at the cloud top, and therefore cause an underestimate of the r_e profile, so the measurement made at cloud top is not representative of an adiabatic cloud (Nakajima et al., 2010). The issue of entrainment is particularly important for cumuliform clouds, due to the lateral entrainment of dry air. Precipitation may also affect the retrieved r_e , as drizzle-sized droplets grow through collisional growth, creating a non-adiabatic bimodal droplet distribution. However, several studies have found the presence of drizzle does not seem to create significant biases in r_e between satellites and in situ measurements for low-level clouds, particularly relative to the biases from 3D radiative transfer or cloud heterogeneity effects (e.g., Painemal and Zuidema, 2011; Fu et al., 2022). These results are also in agreement with theoretical studies (Zinner et al., 2010; Zhang et al., 2012).

2.1.3 N_d and LWP

The biases and uncertainties associated with the r_e and τ_c retrievals contribute to uncertainties in the derived cloud properties, N_d and LWP. Assuming an adiabatic cloud (one which is non-precipitating, does not interact with its environment through processes such as entrainment and has increasing LWP increases with height), N_d and

LWP can be expressed as (Brenguier et al., 2000; Wood and Hartmann, 2006):

$$N_d = \frac{1}{2\pi k} \sqrt{\frac{5}{Q\rho_w}} (f_{ad}c_w)^{\frac{1}{2}} \tau_c^{\frac{1}{2}} r_e^{-\frac{5}{2}} \quad (2.1)$$

and

$$LWP = \frac{5}{9} \rho_w \tau_c r_e \quad (2.2)$$

in which ρ_w is the density of water and Q is the scattering coefficient. As droplets are typically much larger than the NIR wavelengths, the geometrical optics limit is assumed to be met, giving a value of Q approximately equal to 2 (Bennartz, 2007). In Equation 2.2, the factor of $\frac{5}{9}$ arises from assuming an adiabatic cloud with increasing liquid water and r_e profiles with height (e.g., Wood and Hartmann, 2006).

In Equation 2.1, k relates the effective radius to the mean geometrical radius and is associated with the cloud droplet spectrum width. In an adiabatic cloud model, k is assumed to be constant throughout the cloud; however, processes such as entrainment mixing may invalidate this assumption. Painemal and Zuidema (2011) found the droplet spectrum width to narrow with height in the cloud, leading to larger values of k . However, other studies have found little variation of k within clouds (e.g., Brenguier et al., 2011). Across a range of cumulus and stratiform cases, Brenguier et al. (2011) found a range in k of between 0.7 to 0.9, with Martin et al. 1994 finding a narrower range of 0.7 - 0.8 for warm stratocumulus clouds. Bennartz and Rausch (2017) estimate the uncertainty in k due to this range to be around 20%, while Merk et al. (2016) find a smaller uncertainty of 12.5%. In this work, k is assumed to have a constant value of 0.8, as measured as the profile average for marine stratocumulus clouds in Painemal and Zuidema (2011).

The condensation rate (c_w) varies with temperature and pressure (Ahmad et al.,

2013). The pressure dependence is weak, so the cloud top pressure is commonly used to represent the value throughout the cloud (as is done in this thesis), with the errors associated with this approximation typically small (e.g., Grosvenor and Wood, 2014). However, the stronger dependence on temperature may introduce biases associated with the MODIS cloud top temperature (CTT) retrieval. MODIS has been found to underestimate the cloud top temperature relative to other satellite instruments or aircraft measurements (e.g., Tietze et al., 2011; King et al., 2013). Here, the condensation rate is applied using the linear relationship derived in Gryspeerdt et al. (2016) (following Grosvenor and Wood, 2014). An underestimate of CTT would lead to an underestimate of c_w , and therefore N_d .

The effects of the adiabatic assumption on cloud microphysical property retrievals have been discussed in Section 2.1.2. However, due to processes such as mixing in of dry air, real clouds are usually sub-adiabatic. This is accounted for in the N_d calculation by the subadiabatic factor (f_{ad}), which represents the degree to which the cloud follows the adiabatic profile, with a value of 1 indicating a perfectly adiabatic cloud. Several field campaigns or in situ measurements have made estimates of f_{ad} , finding it can vary widely, with aircraft measurements finding values between 0.1 and 0.9 (Grosvenor et al., 2018). The variation depends on cloud type; for example, marine stratiform clouds are generally found to be closer to adiabatic (Zuidema et al., 2005; Brenguier et al., 2011). Even using active sensors, it is challenging to obtain estimates of f_{ad} ; using ground-based radar and ceilometer, Merk et al. (2016) found a mean f_{ad} of 0.63 ± 0.22 . An estimate of f_{ad} can be made from combining MODIS and CALIOP measurements; using this method, Lu et al. (2023) found that f_{ad} decreased with the cloud base temperature due to increased entrainment-evaporation. However, given the small footprint of the active sensors, these estimates are not available for all of the clouds seen by MODIS. Due to these uncertainties, f_{ad} contributes significantly to the uncertainty in calculating N_d , representing 30% of the total error

(Merk et al., 2016). The work presented here uses an f_{ad} of 0.7, derived by Painemal and Zuidema (2011) for southeast Pacific stratocumulus clouds.

From Equation 2.1, it can be seen that N_d is most sensitive to r_e (raised to the power of $-\frac{5}{2}$). Therefore, the uncertainties in r_e discussed in Section 2.1.2 can introduce significant biases in N_d . In addition to the uncertainties associated with the adiabatic assumption, there may be biases introduced due to instrument uncertainty in measuring r_e and τ_c ; these are assessed to be around 10% for both r_e and τ_c (Platnick et al., 2017; Grosvenor et al., 2018). When compounded with the errors discussed in Section 2.1.2, Grosvenor et al. (2018) find the total error in r_e and τ_c to be 27% and 25%, respectively. Considering all of the errors in c_w , f_{ad} and k , Grosvenor et al. (2018) estimate the total error in N_d of 78%, with over 60% of that error arising from the biases in r_e . However, Gryspeerdt et al. (2022b) found a high correlation between MODIS N_d measurements and measurements made from flight campaigns, with r^2 of up to 0.9 in 1° by 1° regions. Here, the MODIS N_d measurements were filtered to reduce uncertainties, such as excluding cases with high SZA or SPI. Additionally, these high correlations were found in stratocumulus regimes, where assumptions such as the adiabatic cloud model may be more accurate. The error in N_d was estimated to be 30-50%, indicating the utility in filtering the MODIS data to reduce uncertainties.

The adiabatic assumption may also introduce biases in the LWP retrieval, as processes such as entrainment may violate the requirement of increasing water content with cloud height in adiabatic clouds. However, comparisons of MODIS-derived LWP calculated using this assumption show strong agreement with microwave satellite measurements, which do not depend on the adiabatic assumption, particularly in marine stratocumulus (e.g., Bennartz, 2007; Borg and Bennartz, 2007); this is discussed more in Chapter 3, including an analysis on the effects of different data filters

on the agreement between MODIS LWP and another satellite instrument which uses a different retrieval algorithm.

Ship-based measurements have found stratus cloud LWP to be strongly correlated to the expected adiabatic profile (e.g., Zuidema et al., 2005). When comparing in situ and MODIS LWP measurements, Noble and Hudson (2015) found a 14% bias between the two measurements for stratocumulus clouds, largely due to MODIS overestimating r_e . Painemal and Zuidema (2011) also found a positive bias of around 12 g m^{-2} between in situ and MODIS measurements, again largely due to discrepancies in r_e . In cloud regimes which are less likely to be adiabatic, such as cumuliform clouds, other assumptions such as a vertically uniform LWP profile may be more appropriate for calculating LWP due to strong entrainment breaking the adiabatic assumption (e.g., Borg and Bennartz, 2007; Zhou et al., 2016; Miller et al., 2016).

2.1.4 Cloud Phase

As discussed in Section 1.1, ice-containing clouds can have very different effects on the Earth’s radiation budget to their liquid-only counterparts. Therefore, correct determination of cloud phase in satellite retrievals is essential. This is particularly true in the Arctic, where mixed-phase clouds are common. Furthermore, because ice particles and cloud liquid droplets have different scattering and absorption properties, an incorrect determination of cloud phase by MODIS would lead to incorrect retrievals of cloud microphysical properties (Figure 2.1). For example, if an ice-containing cloud were misclassified as a liquid cloud, it may create a positive bias in the r_e retrieval, leading to an underestimate of the N_d (Equation 2.1). When compared with CALIOP, an active lidar instrument, the Collection 6 phase algorithm performed well, particularly over ocean surfaces, which had agreements over 90%

(Marchant et al., 2016; Platnick et al., 2017).

The MODIS C6 cloud optical property phase determination algorithm uses a simple majority voting methodology based on a series of tests to determine the cloud phase (Platnick et al., 2014). In its diagnosis, the algorithm uses the 1 km cloud top temperature and the effective radius retrievals as ice and liquid particles have different imaginary refractive indices in the NIR. It also leverages information from the 1.38 μm channels from the MODIS cloud mask, which tests for high cirrus clouds, and the 1 km IR cloud phase, which depends on the differences in ice and liquid emissivity in the IR channels. A 1 km MODIS pixel can be assigned one of three categories; liquid, ice or undetermined. Previous versions of the MODIS optical property phase algorithm included a ‘mixed-phase’ category; however, due to ambiguities in components such as the IR cloud phase data, this was removed and ambiguous retrievals were designated ‘uncertain’ (Baum et al., 2012; Menzel et al., 2015). Many mixed-phase clouds are composed of supercooled-liquid cloud tops with an ice-containing layer beneath them (Morrison et al., 2012), and therefore may appear as liquid-only clouds to a passive sensor, which can only see the cloud top. Khanal and Wang (2018) show that treatment of a mixed-phase cloud as liquid can introduce significant biases in look-up-tables for the microphysical properties (Figure 2.1), and therefore overestimate r_e and τ_c , particularly for clouds with lower optical depths. However, they found that the biases in cloud properties such as LWP introduced by the high SZA were significantly larger than those associated with incorrect phase determinations.

2.2 Reanalysis

Due to the difficult operating conditions, there is a lack of in situ observations of atmospheric variables in the Arctic. Passive satellites are restricted by their inability to retrieve aerosol optical depth (AOD) in cloudy scenes, which are frequent in the Arctic. Although active sensors can retrieve vertically-resolved aerosol profiles, they have limited spatial resolution, and their retrieval algorithms are based on assumptions about aerosol properties that are not necessarily applicable in the Arctic (Tesche et al., 2014). Therefore, many studies which aim to characterise change in the region rely on atmospheric reanalyses data products. These datasets assimilate a wide array of measurements, such as atmospheric soundings and satellite data to provide a continuous, high-resolution ‘best estimate’ of the atmospheric state across the globe. In regions like the Arctic, where direct observations are scarce, reanalysis data becomes indispensable for characterizing environmental changes and facilitating detailed climatological research (e.g., Fletcher et al., 2016a; Papritz and Spengler, 2017; Graham et al., 2017; Rinke et al., 2017; Dahlke et al., 2022). This thesis uses the fifth-generation reanalysis product from the European Centre for Medium-Range Forecasting (ECMWF), ERA5 (Hersbach et al., 2020) to investigate atmospheric conditions in the Arctic.

ERA5 has been assessed through comparisons with in situ observations in the Arctic to determine its accuracy in simulating key atmospheric variables. Graham et al. (2019b) compared several reanalysis estimates of wind speed, specific humidity and temperature to radiosonde measurements over the Fram Strait, finding that ERA5 simulated the observational data well (correlation coefficients 0.83 - 0.98), and often outperformed other reanalysis products. However, ERA5 still struggled to capture specific aspects of the Arctic climate, such as temperature inversions over cold water. Even over sea ice, where reanalysis products typically have struggled (Wesslén

et al., 2014; Wang et al., 2019), ERA5 shows a good correlation with in situ measurements (Graham et al., 2019a). Crucially for research presented later in this thesis, Graham et al. (2019a,b) show that over both ocean and ice the near-surface wind speeds have good correlation with observational measurements from a range of platforms. Additionally, Renfrew et al. (2021) found that ERA5 performed very well against direct observations of variables such as temperature and wind speed made over the ice-free ocean, but struggled over the MIZ zone due to issues with the representation of sea ice in ERA5. While continued evaluation and improvement of ERA5 are needed, these comparison studies indicate that ERA5 can simulate key atmospheric variables, and therefore can be used to represent the Arctic environment in this thesis.

ERA5 data have horizontal grid resolution of 31 km and vertical resolution of 137 model levels, with an hourly temporal resolution. In this thesis, the data were re-gridded to a 25 km by 25 km polar stereographic grid to match the satellite data using bilinear interpolation.

The Copernicus Atmosphere Monitoring Service (CAMS) dataset, also produced by ECMWF, provides reanalysis data for various atmospheric constituents (Inness et al., 2019). Due to the scarcity of aerosol measurements in the Arctic (Schmale et al., 2021), CAMS provides essential data for estimating aerosol conditions in this region. The CAMS AOD product, which assimilates MODIS AOD retrievals, is produced at 3 hourly resolution and around 80 km horizontal resolution. It includes model estimates for various aerosol species concentrations such as dust, sea salt, organic matter, black carbon, and sulfate. While the CAMS AOD product hasn't been as thoroughly evaluated in the Arctic as the ERA5 datasets, Xian et al. 2022 show that CAMS can reproduce AOD climatologies and trends in the region when compared with available satellite and ground-based measurements. However, they also note

discrepancies between reanalysis products, such as differences in speciated AOD, which underscores the challenges in accurately representing aerosol characteristics in the Arctic. The CAMS AOD data used in this thesis are processed in a similar manner to the ERA5 data.

2.3 DARDAR

Due to the difficulties of using the cloud phase data from MODIS, notably in the cases of mixed-phase cloud, phase data for this work is also obtained from DARDAR v2 (raDAR/liDAR; Delanoë and Hogan, 2010; Ceccaldi et al., 2013). DARDAR is a merged data product synthesised using lidar data from CALIOP (Winker et al., 2009) and radar data from CloudSat (Stephens et al., 2008).

CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) is a dual wavelength, polarization-sensitive lidar, which flew in NASA's A-Train alongside Aqua until 2018, allowing for synergistic studies of clouds alongside other satellite instruments. CALIOP provides vertically-resolved cloud layer information, with horizontal and vertical resolutions of up to 333 m and 30 m, respectively. CALIOP uses short wavelengths of light (532 nm and 1064 nm), making the lidar very sensitive to small cloud particles. However, as the lidar is also easily scattered and absorbed by the cloud particles, the signal is rapidly attenuated through multiple scattering and does not penetrate deeply into optically thick clouds. Lidar backscatter is proportional to the second moment of the cloud particle size distribution, meaning it is sensitive to the cloud particle concentration (Donovan and van Lammeren, 2001). Hence, the retrieval is typically dominated by liquid droplets, due to their high number concentration even in ice-containing clouds (Hogan and O'Connor, 2004).

CloudSat is a millimeter wavelength (94 GHz) nadir-viewing cloud radar, which was also in the A-Train orbit until 2018. The data are produced at a 1.4 km cross-track and 1.9 km along-track resolution and, as an active sensor, it allows for the study of the vertical structure of clouds with a 485 m vertical resolution. CloudSat's radar uses much longer wavelengths compared to lidar data, significantly reducing the likelihood of signal attenuation. Therefore, it allows for the analysis of cloud particle properties in deep clouds and can even detect below-cloud precipitation.

As the CloudSat wavelength is typically larger than most detected particles, the retrieved signal is mostly based on the Rayleigh scattering regime, and therefore is proportional to the sixth power of the particle size (Lohmann et al., 2016). As a result, it would only take a relatively small number of large particles (such as ice crystals) in the sampling space to dominate the radar return, even in the case of a small number of ice crystals present with a large number of small droplets (e.g., Hogan et al., 2003; Wang, 2019).

DARDAR exploits the strengths of both CloudSat and CALIOP to provide vertically-resolved information about cloud phase. The phase determination algorithm incorporates data from the CALIOP L1B attenuated backscatter profiles at 532 nm (β_{532}), the CloudSat 2B-GEOPROF radar reflectivity and ECMWF auxiliary (ECMWF-AUX) temperature data. The CALIOP Vertical Feature Mask is also used to give more information about the targets and to distinguish clouds from aerosols (Ceccaldi et al., 2013). Figure 2.2 shows the decision tree logic of the phase classification in DARDAR.

The details of this phase algorithm are outlined in Delanoë and Hogan (2010) and Ceccaldi et al. (2013), and summarized below. β_{532} is used due to its higher sensitivity to large concentrations of smaller cloud droplets than the 1064 nm channel. A

strong backscatter intensity at this wavelength is due to the presence of liquid water, very high concentration of high particles, or some mixture of the two (Ceccaldi et al., 2013). Following the detection of a strongly backscattering layer, if that backscatter intensity is quickly attenuated (namely, drops by a factor of 10 over 480 m), then the cloud is tested using the ECMWF wet bulb temperature and the cloud thickness to determine the phase. As radar cannot detect supercooled-only layers due to its sensitivity to larger particles (Hogan et al., 2003), a radar return signifies that either there is ice also present in the layer (in the case with the ‘ice + supercooled water’ categorisation), or the layer is either ice-only or rain or drizzle are present, based on temperature.

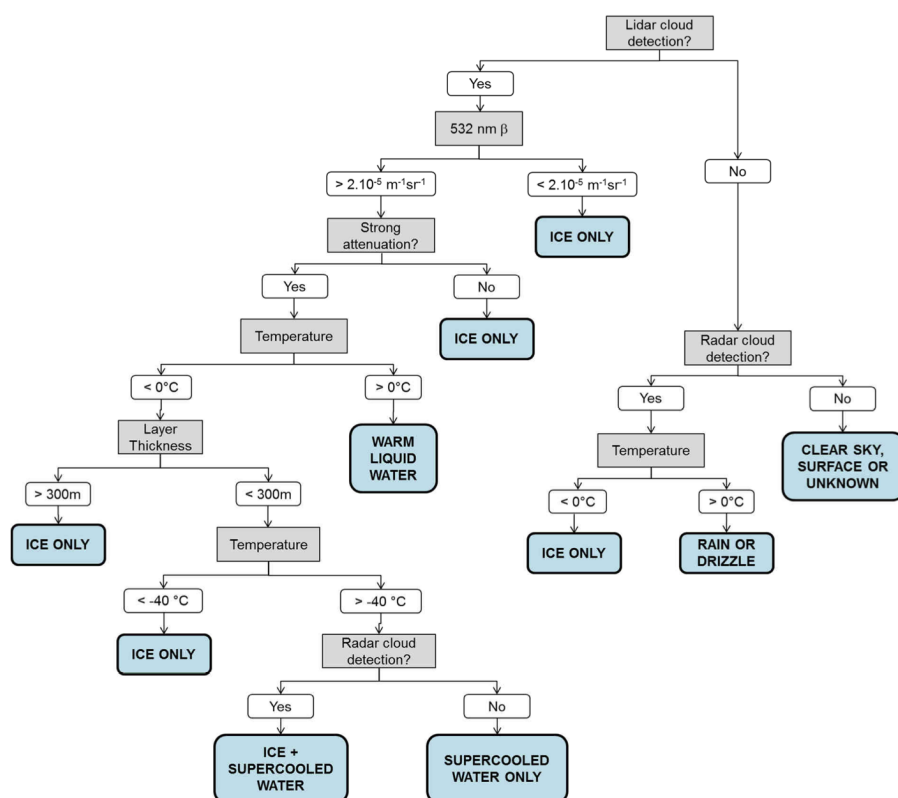


Figure 2.2: Decision tree of the DARDAR v2 phase determination algorithm. Taken from Mioche et al. (2015).

As the lidar data is produced at a higher resolution than CloudSat, several CALIOP

retrievals are matched to each radar retrieval and the radar is oversampled vertically when merged, resulting in a vertical resolution of 60 m and a horizontal resolution of 1.1 km. While DARDAR can harness the strengths of both lidar and radar, it is also vulnerable to the weaknesses in both datasets. For example, the lowest-altitude radar retrievals can suffer from ground clutter due to reflections from the surface, creating a blind zone (Marchand et al., 2008). This can lead to CloudSat, and therefore potentially DARDAR, biasing the retrievals of the lowest-level cloud cover (Marchand et al., 2008; McErlich et al., 2021). Similarly, attenuated lidar retrievals can lead to underestimates in cloud cover (Silber et al., 2018). Relative to in situ measurements, these problems with the satellite can create negative biases of up to 25% in DARDAR cloud cover relative to ground-based observations between the surface and 500 m (Blanchard et al., 2014). However, DARDAR cloud occurrence typically agrees well with in situ measurements (Mioche et al., 2015).

When DARDAR is used in this work, the phase fraction for each profile is calculated as the number of DARDAR retrievals for each flag divided by the number of DARDAR retrievals for each 25 km by 25 km grid box (to match the MODIS data). For this thesis, the ‘ice’ category comprises DARDAR phase flags ‘Ice’, ‘Spherical_or_2D_ice’ and ‘Highly_concentrated_ice’ and DARDAR flag ‘Supercooled_and_ice’ is considered ‘mixed-phase’. The method of defining mixed-phase profiles in DARDAR from Danker et al. (2022) is used here; if a ‘mixed’ layer is above an ‘ice’ layer, a ‘supercooled’ above ‘ice’, any combination of ‘supercooled’ and ‘mixed’ or any combination of ‘supercooled’ or ‘mixed’ above ‘ice’, then the layer is considered ‘mixed’. The ‘warm liquid’ and ‘supercooled liquid’ DARDAR categories are combined to create a single ‘liquid’ category. Multi-layer clouds are excluded by removing profiles which have more than three ‘clear sky’ or failed retrievals (equating to a distance of 240 m) between cloud layers, as in previous work (Danker et al., 2022).

Chapter 3

Assessing Biases in MODIS Liquid Water Path Retrievals Through Comparison with AMSR-E

Key Points

- The MODIS cloud top property algorithm is limited under specific conditions, leading to biases in cloud property measurements.
- The most significant discrepancies between AMSR-E and MODIS data arise from misclassified ice clouds, undetected sea ice, and high solar zenith angles.
- Applying strict filters to MODIS data significantly enhances its agreement with AMSR-E measurements.
- The AMSR-E algorithm's threshold for partitioning liquid water and rainwater is set too low, introducing additional bias in LWP estimates.

3.1 Introduction

As explored in Section 2.1.2, the LWP exerts a significant influence on cloud albedo, which in turn impacts the shortwave cooling effect. Consequently, reliable satellite measurements of LWP are critical for determining the influence of aerosols on clouds and their subsequent effect on climate. The MODIS satellite instrument provides state-of-the-art cloud microphysical property measurements, which are key to estimating LWP. However, as highlighted in Section 2.1, there are certain conditions in which the assumptions underpinning the MODIS algorithm may become invalid. Understanding the limitations of the MODIS retrieval is essential for using these data to improve our understanding of cloud-related processes.

The Advanced Microwave Scanning Radiometer for EOS (AMSR-E) ocean product is often used to identify the effects of approximations such as the adiabatic cloud model on MODIS LWP (Bennartz, 2007; Greenwald, 2009; Seethala and Horváth, 2010; Lebsock and Su, 2014; Khanal and Wang, 2018). Its determination of cloud liquid water path is based on a different set of physical principles (Wentz and Meissner, 2004). Passive microwave instruments retrieve the cloud LWP from the observed brightness temperature, with the retrieval based on 37 GHz channel due to its sensitivity to condensed liquid water. Additionally, at this frequency the emission signature of liquid is an order of magnitude larger than that of ice particles (Battaglia et al., 2006), so the LWP retrievals are largely unaffected by the presence of ice. Since this method relies on the emission and absorption, rather than the scattering, of solar radiation, it is not susceptible to errors associated with 3D radiative effects or inaccuracies arising from the plane parallel assumption.

In general, there is a strong agreement between AMSR-E and MODIS LWP in high coverage, warm, non-precipitating stratocumulus clouds (correlation coefficients be-

tween 0.89 - 0.96; Borg and Bennartz 2007; Bennartz 2007; Seethala and Horváth 2010; Lebsock and Su 2014). These conditions align closely with the adiabatic profile and plane-parallel assumptions, reducing biases in MODIS data. Globally, AMSR-E overestimates MODIS LWP, except at high latitudes, where MODIS LWP can be significantly higher than the AMSR-E product (e.g., Greenwald, 2009; Seethala and Horváth, 2010; Lebsock and Su, 2014; Zhou et al., 2016). Seethala and Horváth (2010) suggested that the MODIS underestimation at lower latitudes could be partly due to non-adiabatic processes like entrainment, particularly in cumuliform clouds. This would reduce r_e and therefore the LWP (based on Equation 2.2). The discrepancy at high latitudes can be attributed to biases in the MODIS data during low sun conditions; above a solar zenith angle of 65° , the positive MODIS bias can reach 50% (Greenwald, 2009; Lebsock and Su, 2014; Zhou et al., 2016), with greater differences for more heterogeneous clouds due to 3D radiative effects (Seethala and Horváth, 2010). The positive bias MODIS LWP is due to the significant increase in τ_c with SZA (Khanal and Wang, 2018), discussed in Section 2.1.2.

MODIS data underpin a significant amount of the analysis presented in this thesis. Therefore, understanding the ability of MODIS to accurately retrieve cloud properties is key for establishing confidence in the subsequent results. This study will compare MODIS with AMSR-E to determine the conditions in which the MODIS cloud microphysical retrievals are reliable and justify the use of MODIS throughout this work, despite the difficult Arctic conditions.

3.2 Methods

Previous studies have addressed biases in MODIS by selectively using data from scenarios where the assumptions of the microphysical retrieval algorithm remain valid

(e.g., Gryspeerdt et al., 2019a). Here, I investigate the effect that applying such filters to the MODIS LWP data has on the agreement between the two instruments. Data from 2008-2011 were included in the analysis. This period is shorter than the remainder of the thesis due to a mechanical failure in AMSR-E in October 2011 which prevented further data collection. The study is limited to using data from the MODIS sensor on the Aqua satellite, to match temporally with the AMSR-E sensor, also aboard Aqua. The AMSR-E data were regridded from their native resolution of 12 km by 12 km to match the MODIS grid. The analysis was performed on an orbital level to prevent the effects that temporal averaging may have on the results. Only pixels above 60° latitude were included in this work.

The MODIS LWP is calculated only using pixels retrieved as ‘liquid’ according to the MODIS ‘Cloud_Phase_Optical_Properties’. However, this may mean that in each aggregated 25 km by 25 km grid box, not all of the pixels included are actually used in the LWP calculation. The potential biases introduced by these non-liquid pixels are considered later in this chapter. There may also be some additional uncertainty if only a small number of the valid pixels remain (i.e., very few of the 625 pixels input into the grid were retrieved as liquid). Additionally, as deriving the MODIS LWP is dependent on the microphysical properties (following Equation 2.2), which in turn are only retrieved when incoming solar radiation is present, the analysis is effectively restricted to polar day (approximately March to October, dependent on the latitude). AMSR-E is not reliant on solar radiation, so these data are restricted to points where there are valid MODIS microphysical retrievals. As the AMSR-E LWP is derived across the whole scene, including cloud-free areas, the in-cloud AMSR-E LWP is calculated by dividing the AMSR-E LWP by the MODIS liquid cloud fraction (following Bennartz, 2007).

For MODIS, some basic filters were applied, including removing multi-level clouds

(MODIS ‘Cloud_Multi_Layer_Flag’) and only including liquid-phase 1 km by 1 km pixels (‘Cloud_Phase_Infrared’, ‘Cloud_Phase_Optical_Properties’ flags). The sea ice pixels were removed using daily sea ice cover data from Nimbus-7 SMMR and DMSP SSM/I-SSMIS Passive Microwave Data, Version 2 dataset (DiGirolamo et al., 2022), which are at a 25 km by 25 km resolution. The other filters to be investigated and the justification for doing so are included in Table 3.1 and discussed in Section 2.1.

AMSR-E’s measurement of microwave brightness temperature integrates key environmental factors – sea surface temperature, water vapor column, and sea surface wind speeds – into its LWP calculations. However, errors in the retrieval of these quantities may bias the AMSR-E LWP (e.g., Lebsock and Su, 2014). AMSR-E also cannot distinguish between condensed liquid in the form of rain or cloud droplets, and the retrieval partitions the LWP into rain or cloud water based on an empirically derived threshold of 180 g m^{-2} . This assumption can introduce a bias into the cloud LWP by assigning too great a portion of the signal to rain in thicker clouds (e.g., Lebsock and Su, 2014; Greenwald et al., 2018). Biases can arise from the cloud top temperature derived by AMSR-E using the sea surface temperature and water vapour column, as colder clouds have a stronger microwave absorption (e.g., Seethala and Horváth, 2010). The microwave retrievals may over-estimate cloud fraction in clear skies or broken cloud scenes due to inaccurate assumptions of surface emissivity, known as the ‘clear-sky’ bias (Greenwald 2009; Seethala and Horváth 2010).

3.3 The LWP climatologies

Figure 3.1 shows the LWP climatologies generated from the MODIS and AMSR-E instruments for 2008-2011, inclusive. Figure 3.1 (a) and (b) shows the MODIS and AMSR-E LWP with only the ‘basic’ sea ice filter, with (c) showing the difference be-

Table 3.1: Justifications for MODIS Data Filters

Filter	Justification	Reference
Sea ice	Microwave instruments struggle to retrieve LWP over sea ice	e.g., Haggerty et al. (2002)
Sea ice + 25 km sea ice buffer	Appears that some pixels still have undetected sea ice in AMSR-E LWP maps	Figure 3.1
Low cloud SPI (below 30)	Plane-parallel assumption breaks down for heterogeneous clouds	Zhang and Platnick (2011), Seethala and Horváth (2010)
Low solar zenith angle ($<65^\circ$)	Introduction of 3D radiative effects	Grosvenor and Wood (2014), Khanal and Wang (2018)
Exclude $\tau_c < 4$ and $r_e < 4 \mu\text{m}$	Non-orthogonal retrievals	Quaas et al. (2006)
MODIS CF > 90	MODIS retrievals less accurate at cloud edges	e.g., Marshak et al. (2006)
Removing all ice clouds	Misclassifying liquid and ice introduces biases in the microphysics retrievals	Khanal and Wang (2018)
Non-precipitating ($r_e < 15 \mu\text{m}$)	Issues with AMSR-E rain to cloud water partition	e.g., Lebsock and Su (2014)

tween them. The MODIS LWP has a regional mean 128 g m^{-2} , and shows a high bias relative to the microwave instrument, with an average bias of 25 g m^{-2} but as high as 80 g m^{-2} in the Barents and Kara Sea. This strong MODIS overestimate is in agreement with other studies which show an increase in MODIS LWP at high latitudes when additional filters are not applied to the MODIS data (e.g., Greenwald, 2009). The AMSR-E-derived LWP is generally more consistent with other (non-MODIS) satellite or in situ studies, with a mean of 103 g m^{-2} and a regional mean of 88 g m^{-2} in the Barents and Kara Sea (e.g., Lenaerts et al., 2017). However, some pixels show very mean high LWP (above 250 g m^{-2}). These pixels are concentrated around the sea ice edge, indicating that although a strict binary mask for sea ice is used (any amount of ice detected by the NSIDC product is classified as a sea ice pixel), there may be some undetected ice in the sea ice product. This could lead to abnormally high AMSR-E LWP retrievals and contribute to a significant bias in the mean.

In light of this, a more conservative sea ice mask is created, which includes an additional 25 km ‘buffer’ around the original sea ice mask. This buffer is justified further due to the relatively coarse resolution of the sea ice product (25 km), which introduces uncertainty about the actual location of sea ice edge and marginal ice zone (Cavalieri et al., 1996; Di Girolamo et al., 2010). Figure 3.1 (d) and (e) show the MODIS and AMSR-E LWP with this additional filter. The MODIS LWP is not substantially affected by the stricter filter (mean 131 g m^{-2}). In contrast, the mean AMSR-E LWP has been reduced to 92 g m^{-2} , as many of the particularly high LWP pixels in Figure 3.1 (b) have been removed. This further has the effect of removing the large negative values in Figure 3.1 (f), increasing the mean difference between MODIS and AMSR-E LWPs to 40 g m^{-2} across the region.

Due to the substantial biases introduced by undetected sea ice to the AMSR-E LWP

product, the remainder of this LWP comparison work uses the more conservative sea ice mask (the basic mask plus a 25 km buffer) to ensure a more accurate comparison between the two products.

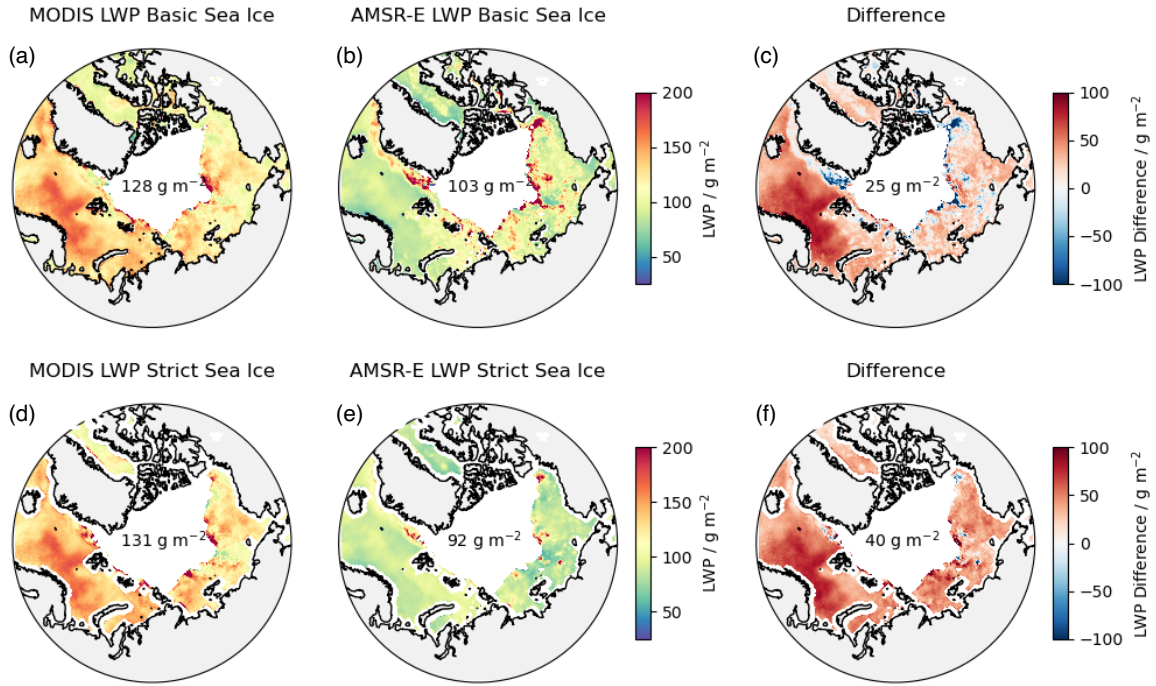


Figure 3.1: Cloud liquid water path climatologies for marine clouds between 2008-2011. (a) and (b) are the derived from the MODIS and AMSR-E products and the basic sea ice filter, and (c) is the difference between them. (d) and (e) are for MODIS and AMSR-E again, but with an additional 25 km buffer for the sea ice mask, and (f) is the difference between them.

3.4 The effects of filters on the LWP climatologies

The filters presented in Table 3.1 were used in processing the MODIS LWP, in addition to applying the strictest sea ice mask. The impact of these filters on the discrepancy between MODIS and AMSR-E LWP is illustrated in Figure 3.2. Each of the filters have different degrees of efficacy - notably, the cloud heterogeneity ($SPI < 30$; Figure 3.2 (b)), the cloud fraction ($CF > 90$; Figure 3.2 (e)) and the precipitation (3.2 (f)) filters do little to improve the agreement between MODIS and AMSR-E, with

mean biases of 44 g m^{-2} , 43 g m^{-2} and 38 g m^{-2} , respectively. The microphysics filter ($\tau_c < 4$ and $r_e < 4 \text{ }\mu\text{m}$; Figure 3.2 (d)) actually increases the MODIS LWP bias to 60 g m^{-2} . It should be noted that while the non-precipitating cloud filter applied to the MODIS data is based on droplet size, using the commonly-applied threshold of a mean droplet effective radius of $15 \text{ }\mu\text{m}$ to denote precipitating clouds (e.g., Rosenfeld and Gutman, 1994), AMSR-E uses a LWP threshold to partition raining and non-raining clouds. Additionally, this filter only requires a majority (more than 75%) of the r_e for each grid box to be smaller than the precipitation threshold to be kept; a minority of raining clouds may bias the results. Therefore, while a useful proxy, this filter may not fully address biases introduced by precipitation.

Two filters significantly reduce the MODIS-AMSR-E bias - restricting the solar zenith angle to below 65° (Figure 3.2 (a)) and excluding all grid points that have any non-zero ice fractions (Figure 3.2 (e)), which reduce the bias to 11 g m^{-2} and 17 g m^{-2} , respectively. The substantial reduction in the MODIS LWP due to the SZA and the ice cloud filters are expected from previous work (Lebsock and Su, 2014; Khanal and Wang, 2018).

Figure 3.3 (a) shows the effects of applying all of the filters to the MODIS data. Relative to Figure 3.2 (a) and (d), it is clear that the mean LWP has been significantly reduced, with a regional mean of 90 g m^{-2} . This reduction in MODIS LWP produces a much better agreement with the AMSR-E LWP, as shown in Figure 3.3 (c). The mean bias between the two instruments has been reduced to -6 g m^{-2} , but it is evident that this masks some variation regionally. There are several regions in which AMSR-E overestimates MODIS (average bias of the negative regions -15 g m^{-2}), in agreement with what is typically seen at lower latitudes in global studies. This illustrates the importance of removing cases in which it is known that the MODIS retrieval struggles - such as in low sun conditions - to obtain reliable retrievals of

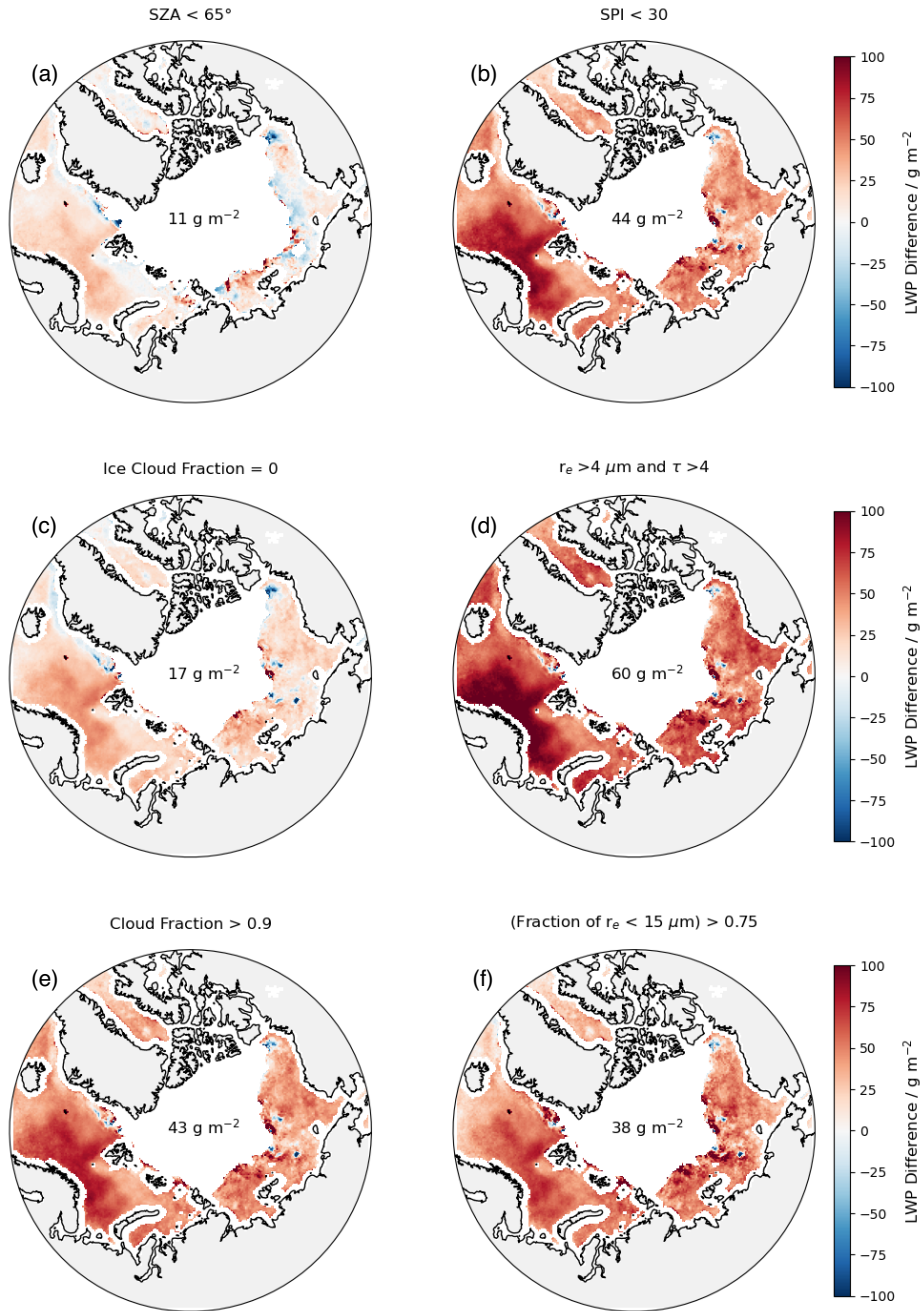


Figure 3.2: Effects of the different filters individually from Table 3.1 on the bias between the MODIS and AMSR-E LWP. All filters have been applied with the stricter sea ice filter. (a) SZA < 65° (b) Cloud heterogeneity (SPI) < 30 (c) Zero ice cloud fraction for the grid box (d) $r_e > 4 \mu\text{m}$ and $\tau_c > 4$ (e) cloud fraction above 0.9 and (f) 75% of r_e retrievals are below the precipitation threshold (15 μm).

cloud microphysical properties.

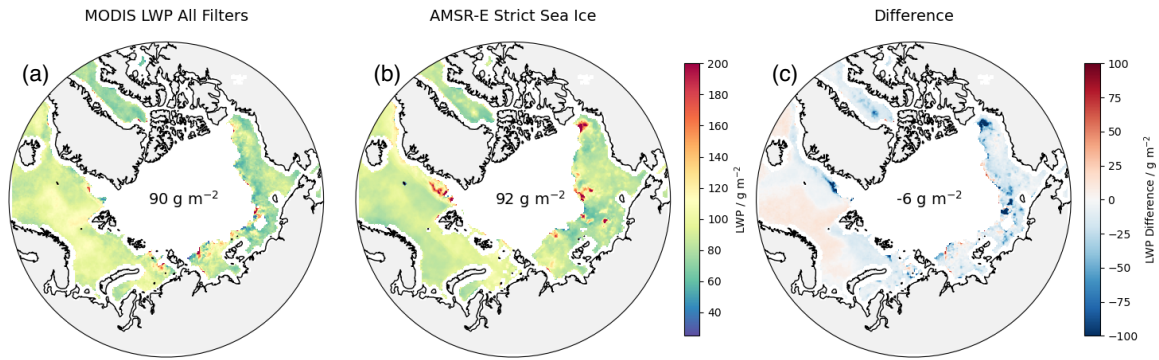


Figure 3.3: As Figure 3.1, but applying all of the filters to the MODIS data (a), the strict sea ice filter to the AMSR-E data (b), and the difference between them (c).

3.5 The effects of filters on the correlation between AMSR-E and MODIS

To assess the impact of the filters on the correlation between the two datasets, conditional probability histograms were created, shown in Figure 3.4. It can be seen that even without refining the data, there is a reasonable correlation between the AMSR-E and the MODIS LWP (0.60 and 0.62 for the basic and strict sea ice filters, respectively). Generally, MODIS tends to overestimate compared to AMSR-E, as observed in the climatology analyses, except at low LWP values where AMSR-E shows a slight overestimation. This AMSR-E overestimation at low LWP may be due to the AMSR-E clear-sky bias, which is when AMSR-E overestimates LWP in clear sky or tenuous cloud conditions due to errors in the water vapour or wind speed retrievals (Lebsock and Su, 2014). It should be noted that these histograms are confined to LWP values below 400 g m⁻² for clarity, thereby excluding the extreme AMSR-E overestimates related to undetected sea ice. However, these values are still considered in the correlation calculations.

The impact of the filters shown in Figure 3.4 is generally in agreement with the results described in Section 3.4. The cloud heterogeneity and the microphysics fil-

ters (Figure 3.2 (d) and (f)) actually reduce the correlation between the two LWP products, relative to the sea ice-only filters. In the case of the microphysics filter, this may be due to removing some low-LWP cases, where there is generally a better agreement between AMSR-E and MODIS. Similarly, restricting the comparison to high cloud fraction or non-precipitating regimes (Figure 3.2 (g) and (h)) offers either no or moderate improvement.

As with the climatologies, the agreement between MODIS and AMSR-E LWP measurements significantly improves following the implementation of SZA and ice cloud filters. This improvement is reflected in the correlation values, which have increased to 0.75 and 0.76 (Figure 3.2 (c) and (e)). The elimination of instances where MODIS significantly overestimates LWP – indicated by the removal of data points from the bottom-right sector of the histogram – contributes substantially to this improved correlation.

Figure 3.5 shows the effects of applying all of the filters to the MODIS data. The correlation increases to 0.89, showing excellent agreement between the MODIS and AMSR-E LWP products. As before, this strong correlation arises from removing many cases in which there are substantial positive biases in the MODIS LWP relative to AMSR-E (the red in the lower-right of Figure 3.5 (c)). These results are in agreement with values typically seen in ‘ideal’, stratiform, non-precipitating, homogeneous liquid scenes at lower latitudes, indicating that the disagreement seen in previous studies was due to the breakdown of certain assumptions in the MODIS algorithm. However, it should be noted that there is still evidently an overestimate in the MODIS LWP at high values. This is explored further in Section 3.5.1.

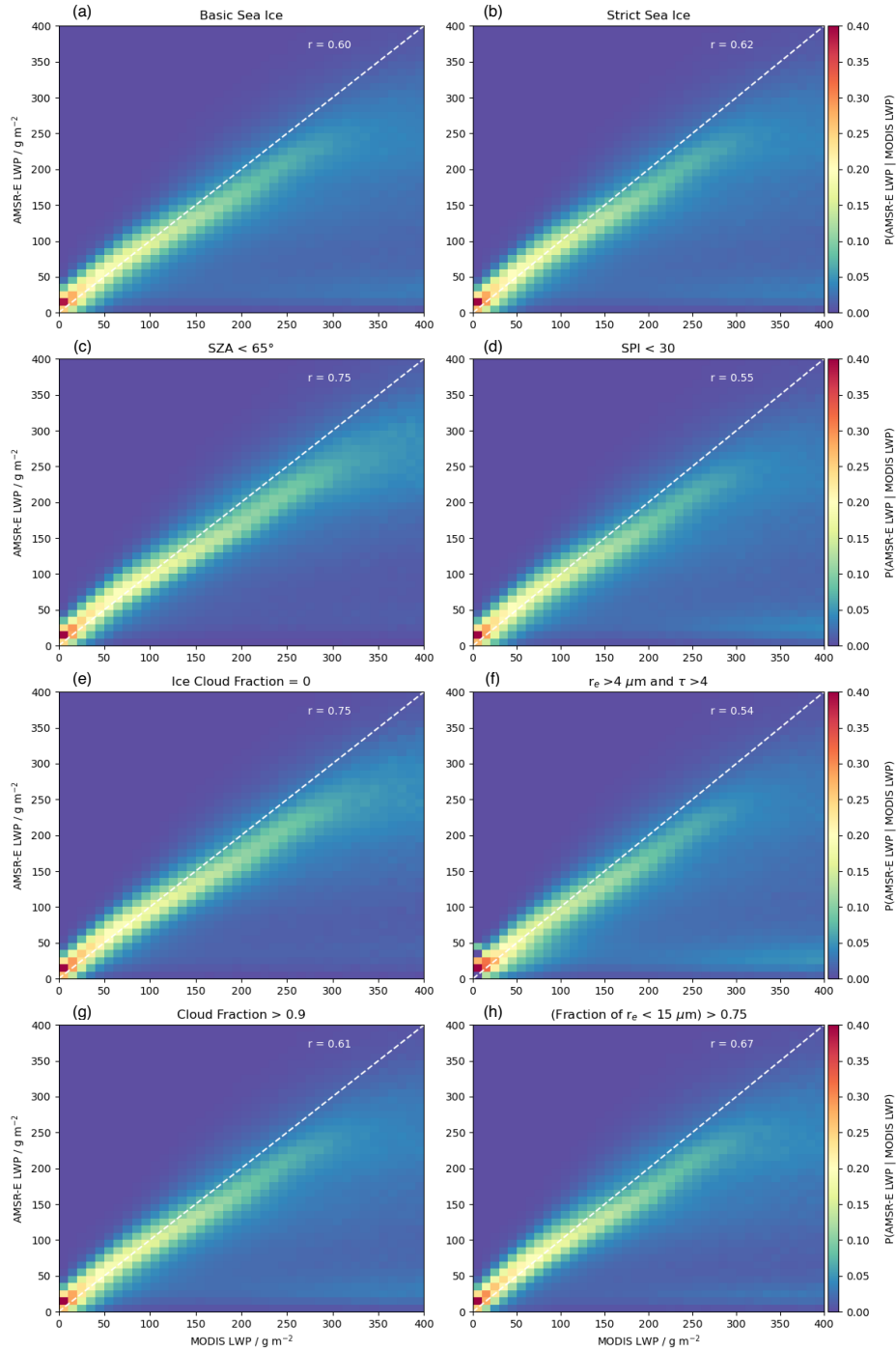


Figure 3.4: Joint probability histograms of AMSR-E and MODIS LWP for the filters described in Table 3.1; (a) basic sea ice, (b) strict sea ice, (c) $\text{SZA} < 65^\circ$ (d) Cloud heterogeneity (SPI) < 30 (e) Zero ice cloud fraction for the grid box (f) $r_e > 4 \mu\text{m}$ and $\tau_c > 4$ (g) cloud fraction above 0.9 and (h) 75% of r_e retrievals are below the precipitation threshold ($15 \mu\text{m}$). The correlation coefficients are shown in the top-right of each plot, and the white dashed line shows the 1:1 correspondence line.

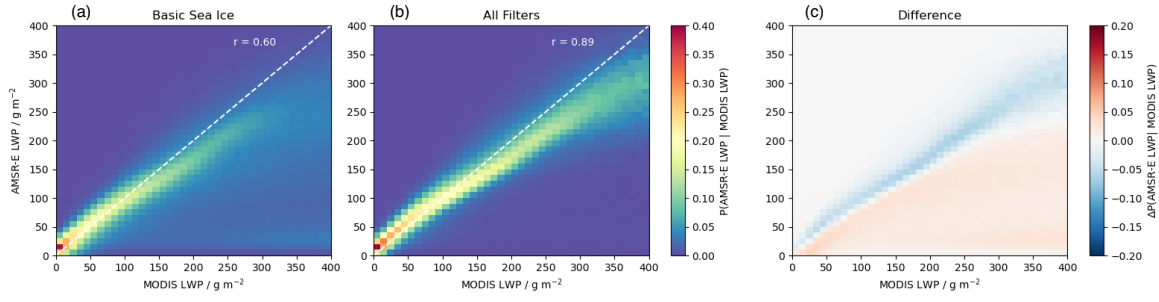


Figure 3.5: Joint probability histograms of AMSR-E and MODIS LWP for (a) the basic sea ice filter, (b) all filters described in Table 3.1, and (c) the difference between them.

3.5.1 Bias as a function of SZA and ice fraction

The evidence so far suggests issues with the retrieval algorithm in low sun and the incorrect retrieval of ice as liquid are the dominant drivers of the divergence between the MODIS and AMSR-E LWP. Here, I consider the effects of each of these factors on the LWP in more detail. In each case, all of the filters except the filter being tested are applied to the data.

Figure 3.6 (a) shows the mean r_e and τ_c as a function of SZA. The mean r_e shows great variability at low SZA, but as there are relatively few points at these angles, this effect is unlikely to bias the total results significantly. Between 40° and 60°, the mean r_e increases by 1.5 μm , after which the r_e slightly decreases. This pattern of increasing and then decreasing r_e is similar to what is observed in Grosvenor and Wood (2014) (their Figure 7), but of a different magnitude; Grosvenor et al. (2018) only see a moderate increase of 0.5 μm until 60°, and a more substantial decrease of 1.5 μm at higher angles, which is effectively the reverse of the trend seen here. In contrast, the τ_c remains relatively steady at low SZA and only significantly increases above around 70°, which is comparable to Grosvenor and Wood (2014) (their Figure 5). The degree of variation across the SZA range is greater for τ_c than r_e , with a near-doubling in the mean τ_c between 70° and 80°. This much greater dependence of the τ_c on SZA is in agreement with previous studies (e.g. Khanal and Wang (2018), their

Figure 8).

Figure 3.6 (b) shows the bias in the LWP as a function of SZA. At low SZA, there is a moderate bias of between -10 and 12 g m^{-2} . Above 60 - 65° , the bias rapidly increases with SZA, increasing by a nearly a factor of seven between 60° and 80° . This mean bias in MODIS LWP relative to AMSR-E of over 100 g m^{-2} at high SZA is in agreement with other studies (e.g., Seethala and Horváth, 2010). Grosvenor and Wood (2014) suggested using a cutoff of 65° to remove the effects of high SZA on the MODIS microphysical property retrievals, to balance the magnitude of the bias with maintaining a statistically significant amount of data. While Figure 3.6 (b) shows that this removes the most significant biases in the LWP while still retaining a significant number of the points (approximately 75%), smaller biases remain at small SZAs.

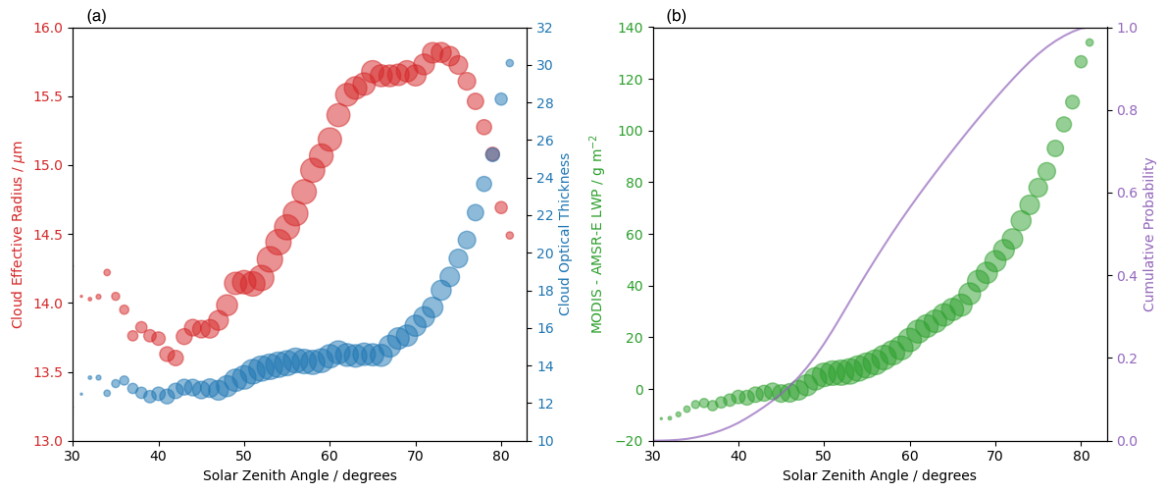


Figure 3.6: Variation in the (a) mean r_e (red) and mean τ_c (blue) and (b) mean difference between MODIS and AMSR-E LWP (green) with solar zenith angle. The cumulative frequency plot is shown purple in (b). The size of the circles is indicative of the number of samples at each point.

Similarly, Figure 3.7 (a) shows the retrieved r_e and τ_c as a function of the ice cloud fraction in the 25 km by 25 km aggregated grid box, and (b) shows the LWP bias and the cumulative frequency. The majority of cases have zero ice fraction and therefore low LWP bias (8 g m^{-2}). However, once ice is present, both r_e and τ_c increase rapidly,

with a corresponding increase in LWP bias of up to 200 g m^{-2} in the most extreme cases. These results are similar to Khanal and Wang (2018); although they looked at the IWP, they also found that more ice leads to significant LWP biases. However, given these cases are relatively uncommon overall, the non-zero ice grid boxes could be removed and still leave a significant portion of the data.

The relationships shown in Figures 3.6 and 3.7 suggest that some sort of bias correction could be applied to the data to account for the variation with ice or solar zenith angle. However, developing such a bias correction algorithm is beyond the scope of this work.

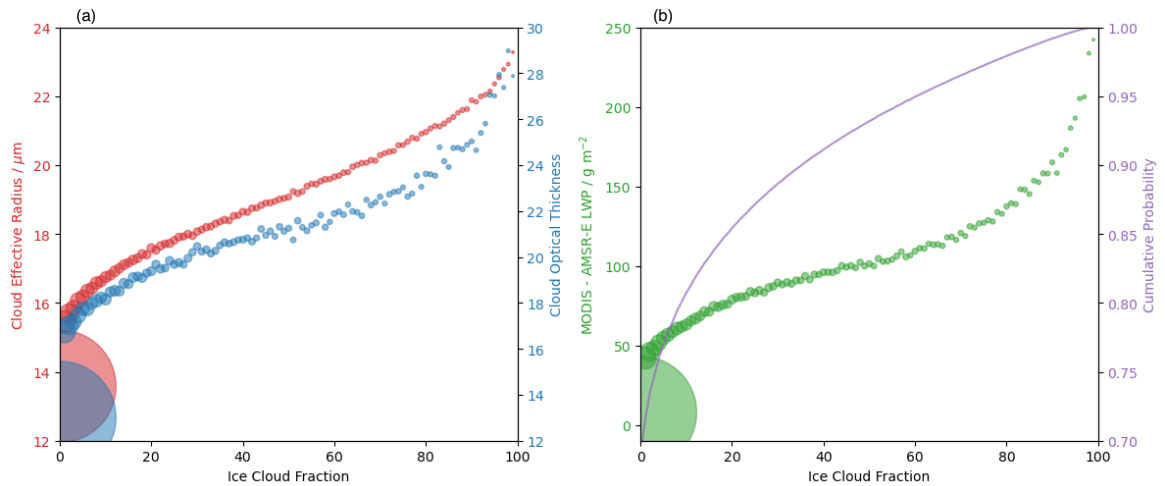


Figure 3.7: As Figure 3.6, but for the ice cloud fraction in each 25 km by 25 km grid box.

3.5.2 Effects of the rain partitioning threshold

So far, this work has only considered filters applied to mitigate issues with the MODIS retrieval algorithm. However, the AMSR-E retrievals are not infallible. For example, as the microwave instrument itself cannot distinguish between condensed water in the form of cloud water or precipitation, the Wentz algorithm used for AMSR-E al-

locates a fraction of the LWP above 180 g m^{-2} to rainfall. This global threshold was established based on studies of extratropical cyclones in the northeastern Pacific (Wentz and Spencer, 1998). Previous work has found that this partitioning may contribute to the observed positive MODIS bias seen at higher LWP. In these cases, thicker, non-precipitating clouds may have a significant portion of their LWP misclassified as rain (e.g., Lebsock and Su, 2014).

Figure 3.8 (orange) shows the average AMSR-E LWP as a function of the filtered MODIS LWP. Below the AMSR-E rain threshold, there is a strong agreement between the two datasets, consistent with earlier findings (Seethala and Horváth, 2010). However, above the 180 g m^{-2} threshold, AMSR-E consistently underestimates MODIS. As the data have been filtered to remove cases known to cause significant MODIS overestimation (such as large SZA), the issue is likely to lie with the AMSR-E LWP.

This divergence after the AMSR-E rain threshold points to potential inaccuracies in the Wentz algorithm’s delineation of cloud and rain water. To investigate this, the LWP that would be retrieved by AMSR-E if all clouds were assumed to be non-raining is calculated using the method described in Appendix A.1, and is shown in Figure 3.8 (green).

This adjusted LWP shows much greater agreement between the two instruments above the 180 g m^{-2} , increasing the mean AMSR-E LWP by around 60 g m^{-2} for the thickest clouds. This indicates that this threshold, which is fixed globally, may be set too low for these conditions and contributes to the AMSR-E underestimation for thicker clouds. However, as less than 8% of the data is found above the threshold (Figure 3.8, blue curve), it has a small impact on the average bias between the two instruments. Calculating the difference in the climatologies as in Figure 3.3,

correcting the AMSR-E LWP changes the bias to approximately -7 g m^{-2} (not shown).

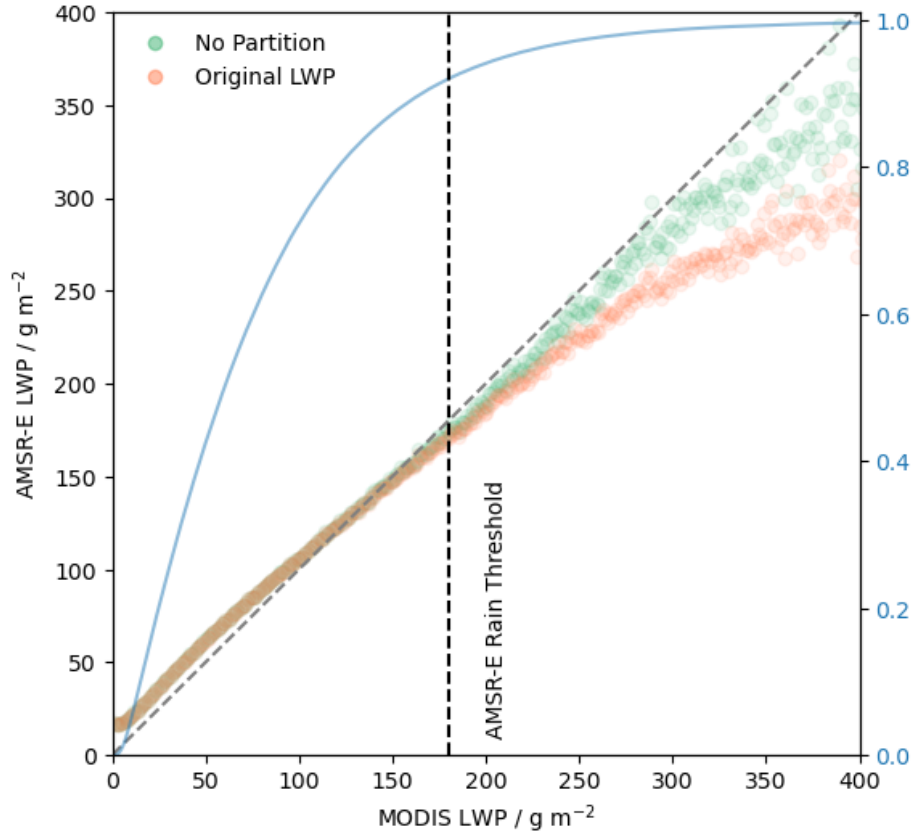


Figure 3.8: Average AMSR-E LWP for each MODIS LWP bin of 1 g m^{-2} for the original data (orange) and the data with the “additional” LWP added (green), and the cumulative frequency plot.

3.6 Discussion and Conclusions

As LWP is a leading control on cloud radiative properties, making reliable observations is crucial to improving our understanding of cloud development and constrain processes such as aerosol-cloud interactions. MODIS provides state-of-the-art retrievals of cloud properties, but these measurements are contingent on assumptions which break down in certain conditions, introducing bias into the LWP measurements. To address this, several studies suggest implementing restrictions on the MODIS data to restrict the data to ‘ideal’ conditions. Here, I investigate the efficacy

of these various filters on improving the reliability of MODIS LWP retrievals through comparison with LWP derived from AMSR-E, a microwave instrument whose retrieval algorithm is based on different physical principals and therefore is not subject to the same biases as MODIS.

The raw MODIS data typically shows a significant positive bias relative to the AMSR-E LWP (Figure 3.1), except in cases of undetected sea ice, when AMSR-E LWP values become very large. However, the unfiltered data still shows a moderate correlation in spite of this positive bias (0.60, Figure 3.4). Investigation into different filtering strategies applied to the MODIS data reveal that a significant portion of this positive bias can be attributed to two situations: the erroneous retrieval of ice-containing clouds as liquid and the breakdown of the plane-parallel assumptions in low sun conditions (Section 3.5.1). Many of the other filters provide little to no improvement on the agreement between the two datasets. For some filters, such as the cloud fraction filter, this is in contrast to previous work; for example, Seethala and Horváth (2010) found much stronger agreement between AMSR-E and MODIS when restricted to high coverage scenes. This may be because their study was global; the Arctic typically has a high cloud fraction (e.g., Curry et al., 1996), so the very low-cloud fraction regimes which show the biggest disagreement between MODIS and AMSR-E are relatively uncommon. While some filters have a relatively small impact on the agreement, their use is justified by our understanding of the limitations of the retrieval algorithm and should be used in future work. When all of the restrictions are applied to the MODIS data, the MODIS and AMSR-E LWP achieve an excellent correlation of 0.89 (Figure 3.5).

While filtering MODIS data significantly improves its agreement with the AMSR-E LWP product, it is crucial to consider the extent of data removal by each filter. If too much data is excluded, the remaining dataset may become too limited to accurately

represent the clouds in the region. Additionally, based on the SZA filtering, a summertime bias would be introduced. Figure 3.9 shows the fraction of data retained by each filter when applied in addition to the strict sea ice filter. More effective filters, such as the SZA, tended to remove a larger portion of the dataset. In contrast, less effective filters, like the SPI filter, excluded a smaller fraction of the data. Applying the SZA filter also introduces a seasonal bias, favouring the inclusion of more summertime pixels when the sun's position is higher in the sky. When all of the filters are applied, only about 40% of the dataset remains. Although the removal of over half of the data poses questions about its representativeness, it is noteworthy that this selective exclusion mainly targets instances where the MODIS retrieval algorithm struggles, therefore justifying the use of a more limited, yet more accurate, dataset.

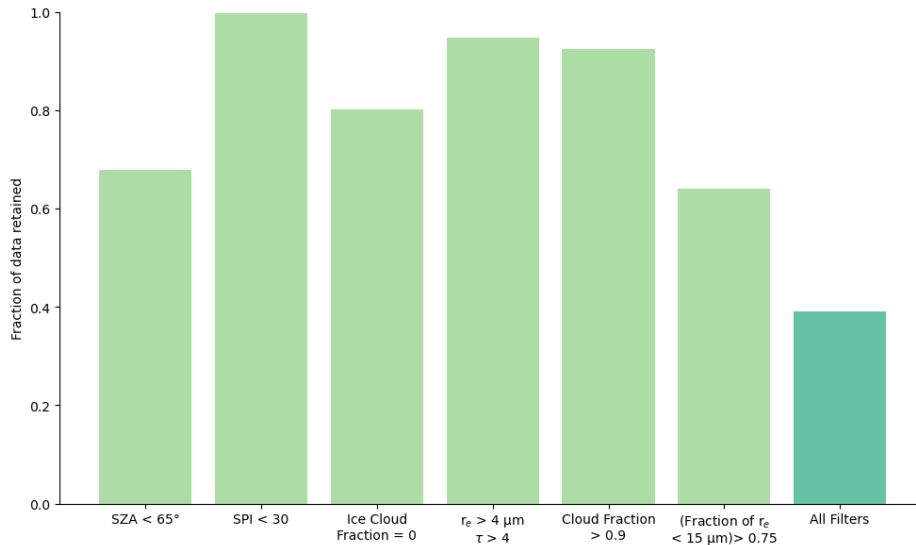


Figure 3.9: Histograms showing the fraction of the data retained following the application of each filter, once the strict sea ice filter had already been applied.

This research highlights the need for caution in using the AMSR-E LWP data, especially due to the strong sensitivity of the microwave instrument to sea ice. Undetected sea ice leads to significant biases, and the implementation of a strict sea ice mask is needed. Additionally, the low threshold at which LWP begins to be converted to rainwater contributes to a MODIS overestimation for thicker clouds. A more re-

alistic threshold is needed, potentially from a wider range of case studies or from global satellite data. However, when AMSR-E is used in Chapter 4, the non-raining LWP correction is not applied. While this correction does improve the agreement between MODIS and AMSR-E, it is in itself inaccurate, as it assumes that none of the clouds about 180 g m^{-2} are raining. Since it is not the goal of this thesis to devise a novel AMSR-E retrieval algorithm, and the correction would only apply to a small proportion of the data, the uncorrected AMSR-E product is deemed sufficient to qualitatively assess the biases in the MODIS data in Chapter 4.

A potential extension of this research could involve comparing satellite-derived LWP with in situ measurements, such as those from the Surface Heat Budget of the Arctic Ocean (SHEBA) campaign (Lin et al., 2003). While these measurements are spatially and temporally limited, their high quality offers a valuable benchmark for assessing MODIS data. They could also allow for a more detailed investigation into the validity of the adiabatic assumption, for example. However, that is beyond the scope of this work.

This study demonstrates that applying specific filters to MODIS data to remove cases where its retrieval algorithm breaks down, significantly enhances the agreement between MODIS and AMSR-E. The most effective filters involve excluding aggregated grid boxes containing any non-liquid clouds and limiting the dataset to low-sun conditions. As noted by Seethala and Horváth (2010), significant changes need to be made to the MODIS retrieval algorithm to address the biases introduced by these conditions. Therefore, filtering the data is a more practical strategy for overcoming these limitations. Although these criteria exclude a considerable fraction of the data, this filtering is justified given the improved quality of remaining data. Overall, the work in this chapter finds that carefully filtered MODIS data has some skill in retrieving cloud LWP under specific conditions, proving its utility in further cloud physics

research.

Chapter 4

Stability dependent increases in liquid water with droplet number in the Arctic

Key Points

- Aerosol-cloud interactions in the Arctic are poorly understood relative to other regions.
- The N_d -LWP relationship shows significant geographical variation; a positive relationship is observed at higher latitudes, in contrast to much of the globe, primarily influenced by lower tropospheric stability.
- There is evidence of a temperature-dependent aerosol effect, or equivalently an aerosol-dependent cloud feedback, due to the dependence on lower tropospheric stability.
- In a warmer and more polluted Arctic, adjustments in LWP could potentially reduce the shortwave cooling effect of clouds.

The contents of this chapter are based on Murray-Watson, R. J. and Gryspeerdt, E.: Stability-dependent increases in liquid water with droplet number in the Arctic, Atmos. Chem. Phys., 22, 5743–5756, <https://doi.org/10.5194/acp-22-5743-2022>, 2022. The analysis and text are my own work, with comments from my co-author.

4.1 Introduction

The relationship between aerosols and Arctic clouds is highly uncertain, in part due to challenges in obtaining reliable observations in the region, as discussed in Chapter 3. However, as industrialisation moves to higher latitudes, understanding how aerosols change cloud properties will become increasingly important (Schmale et al., 2018). This is particularly essential as low-level liquid-containing clouds play a central role in the Arctic energy budget, in which they often contribute to surface heating through their longwave warming effect (Curry and Ebert, 1992; Shupe and Intrieri, 2004). Previous in situ and satellite studies have shown that Arctic clouds are more sensitive to anthropogenic aerosols than their low latitude counterparts (Garrett et al., 2004; Coopman et al., 2018a). The resulting changes in their microphysical properties can substantially change the net radiative effect of the clouds (Lubin and Vogelmann, 2006; Zhao and Garrett, 2015), with the magnitude and sign of the effect also dependent on other factors including season and the albedo of the underlying surface.

Identifying the role of aerosols on cloud properties is further complicated by the influence of confounding variables. For example, increases in satellite-retrieved AOD due to aerosol swelling in high humidity conditions may generate spurious correlations between aerosol and cloud properties (e.g., Quaas et al., 2010). Coopman et al. (2016) found that if meteorology is not accounted for, the magnitude of the cloud response to aerosol is larger by a factor of three than when the confounding

influence of meteorology is reduced by using reanalysis data. To circumvent these issues, some studies (Gryspeerdt et al., 2016) have used a mediating variable, such as N_d . By considering N_d , the relationship between LWP and aerosols can be broken down into two parts; this can be represented using the sensitivity parameter (Feingold et al., 2001), which quantifies the relative change in LWP for a change in AOD (or N_d):

$$\frac{d \ln LWP}{d \ln AOD} = \frac{d \ln LWP}{d \ln N_d} \frac{d \ln N_d}{d \ln AOD} \quad (4.1)$$

Focusing on the N_d -LWP relationship assists in the understanding of the aerosol-LWP relationship as aerosol-induced changes to LWP act via changes in N_d through mechanisms such as precipitation suppression. Previous studies, both globally and in the Arctic, have shown that N_d typically increases with aerosol concentration (e.g. Garrett et al., 2004; Quaas et al., 2006), so the N_d -LWP relationship governs the sign of the LWP response to aerosols. The use of N_d is particularly helpful in the Arctic; the persistently high cloud fraction (Shupe, 2011; Cesana et al., 2012) and high albedo surfaces means passive sensors (such as MODIS) can only obtain limited valid aerosol retrievals. Additionally, N_d is a good choice as its retrieval is not strongly affected by relative humidity.

Although their high temporal resolution and large spatial coverage overcome the issues faced by in situ measurements and field campaigns, few previous studies have used satellites to study Arctic aerosol-cloud interactions, in part due to the difficult conditions for satellites discussed in previous studies (Coopman et al., 2018a; Zamora et al., 2018; Maahn et al., 2021). However, as shown in Chapter 3, carefully filtering the satellite data to the specific conditions can produce reliable data. This work uses several years of passive satellite data to investigate the relationship between LWP and N_d , using reanalysis data to investigate the influence of meteorology.

The findings suggest that the lower tropospheric stability is a dominant control in the N_d -LWP relationship, which may have significant implications in a warmer, ice-free Arctic.

4.2 Materials and Methods

This study uses MODIS data from the Aqua satellite for the years 2010 to 2015, inclusive, using the level 2 collection 6.1 dataset (MYD06_L2; Platnick et al., 2017). The data processing techniques described in Chapter 3 were used on the MODIS data, including application of the filters described in Table 3.1. However, as liquid-topped mixed-phase clouds are common in the Arctic, an additional temperature filter was included to ensure the data were restricted to liquid clouds. Only pixels with cloud top temperatures above 268 K were included in this study, as in situ measurements show these clouds have a liquid water fraction of upwards of 95% (de Boer et al., 2009). MODIS has been shown to underestimate cloud top temperatures in the Arctic (Tietze et al., 2011), meaning that this filtering step is likely removing too many clouds. However, as the inclusion of mixed-phase clouds would introduce significant uncertainties (Chapter 3; Khanal and Wang 2018), this conservative estimate of the cloud top temperature is used. The sea ice mask plus the 25 km buffer were also applied to the MODIS data (Chapter 3).

The cloud liquid water path was estimated according to Equation 2.2, shown again here:

$$LWP = \frac{5}{9} \rho_w \tau_c r_e \quad (4.2)$$

in which τ_c and r_e are the cloud optical thickness and the cloud droplet effective

radius, both acquired from MODIS. Equation 2.2 is discussed in further detail in Section 2.1.3.

As discussed in Chapter 3, comparison with microwave data is useful for identifying potential biases in MODIS. Therefore, LWP data were also obtained from AMSR-E (Wentz and Meissner, 2004). Data from 2010 and 2011 were included in the analysis. The data were processed as in Chapter 3.

The cloud droplet number concentration (N_d) is estimated from MODIS data using Equation 2.1, repeated here:

$$N_d = \frac{1}{2\pi k} \sqrt{\frac{5}{Q\rho_w}} (f_{ad}c_w)^{\frac{1}{2}} \tau_c^{\frac{1}{2}} r_e^{-\frac{5}{2}} \quad (4.3)$$

using parameter values found in Section 2.1.3.

It should be noted that the microphysics filter from Table 3.1 is not applied to the LWP retrievals as N_d is more sensitive to inaccuracies in these values and would introduce a high bias in the MODIS LWP against the AMSR-E LWP (Gryspeerd et al., 2019a). A summary of the filtering methods has been provided in Figure 4.1.

Meteorological reanalysis data was obtained from the ERA5 dataset (Hersbach et al., 2020), processed as described in Section 2.2. The data at the time step which is closest to that of the time of the satellite overpass is considered to be temporally coincident for this study. The effects of free tropospheric moisture and LTS were studied as previous works have shown these variables have a strong influence on the N_d -LWP relationship (Chen et al., 2014; Michibata et al., 2016; Coopman et al., 2016). The specific humidity at 750 hPa (q_{750}) was chosen as a measure of the humidity of the free troposphere. The LTS, which is a measure of the static stability

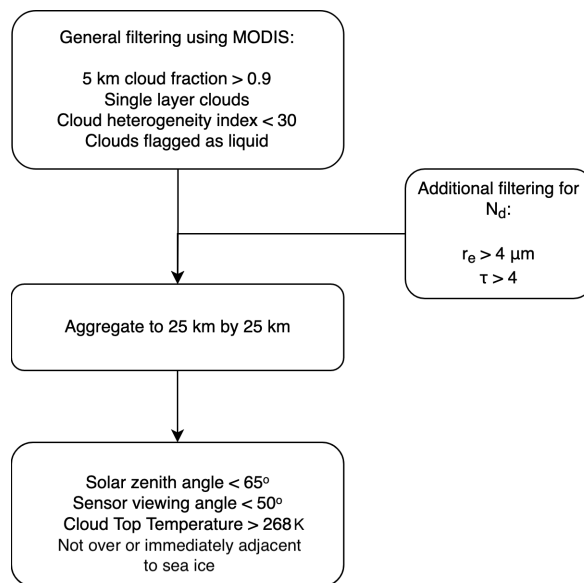


Figure 4.1: Flowchart describing the filtering process used for the MODIS L2 pixels.

of the atmosphere, was calculated as the potential temperature difference between 700 hPa and 1000 hPa (Klein and Hartmann, 1993).

Additionally, the marine cold air outbreak index (MCAO; Kolstad and Bracegirdle, 2008) is important to cloud formation and behaviour at high latitudes (McCoy et al., 2017). Much like LTS, it is a metric of the stability of the boundary layer, although it is calculated as the difference between the potential temperature at 800 hPa and the sea surface temperature (Fletcher et al., 2016a), with positive values indicating higher instability. The MCAO index will be discussed further in Chapter 5.

4.3 Results

4.3.1 The regional and seasonal N_d -LWP relationship

Figure 4.2 (a) shows the annual mean linear sensitivity of the MODIS LWP to N_d . There is a clear negative-to-positive gradient in the sensitivity, with increases in LWP

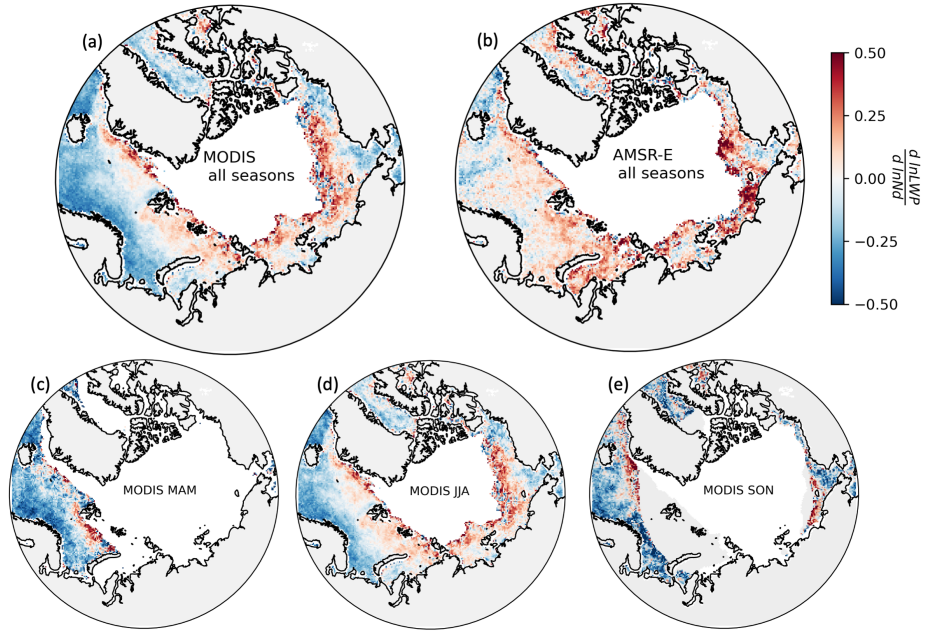


Figure 4.2: The sensitivity of the cloud liquid water path to N_d for (a) MODIS all seasons, (b) AMSR-E all seasons, (c) MODIS March, April and May, (d) MODIS June, July and August and (e) MODIS September, October and November. White indicates data omitted due to the presence of sea ice while grey shows either missing data due to polar night or the presence of land masses.

with N_d typically occurring at higher latitudes. The seasonal cycle of the MODIS sensitivity is shown in Figures 1 (c), (d) and (e). Although spring and autumn have regions of positive sensitivity, the summer months most strongly contribute to the signal observed when considering the all-season response, as they have the most data. Figure 4.2 (b) shows the linear sensitivity between MODIS N_d and AMSR-E LWP for all seasons. The positive relationship and spatial pattern are similar to the MODIS data, with the AMSR-E LWP showing a stronger positive response to aerosols. This may be due to a potential negative bias in the MODIS LWP due to r_e retrieval errors (Gryspeerdt et al., 2019a). Due to the absence of incoming solar radiation during polar nights, there is a lack of data for the winter season, so these months are not shown.

The decrease in LWP with N_d at lower latitudes has previously been observed in

liquid-phase clouds over subtropical oceans (e.g. Michibata et al., 2016; Gryspeerd et al., 2019a) and is consistent with the mechanism of aerosol-enhanced entrainment mixing (Ackerman et al., 2004). Positive sensitivities have previously been observed in high relative humidity conditions (Chen et al., 2014; Toll et al., 2017). Increases in LWP also occur in very clean conditions due to precipitation suppression in this low N_d regime (Gryspeerd et al., 2019a).

There are several possible explanations for the spatial heterogeneity in the LWP response. One potential cause could be due to air masses moving off of the ice edge; cold air moving over the relatively warm open-ocean regions has previously been associated with Arctic cloud formation (Pithan et al., 2018). However, Chapter 5 shows that during these events there is a negative N_d -LWP close to the ice edge. This is in contrast to what is seen here, suggesting that this is not a significant influence on the N_d -LWP relationship. The remaining potential drivers include the cloud mean state and meteorological conditions, which are investigated further in this work.

4.3.2 The role of meteorology and cloud mean state

To investigate the drivers of the different LWP responses, Figure 4.3 shows the distributions of cloud microphysical properties and meteorological variables for grid boxes with positive and negative sensitivity. There is no evidence for a statistically significant difference between the distributions of the LWP, specific humidity, N_d vertical velocity or surface wind speed for two regions (Mann Whitney U test, $p > 0.05$; Mann and Whitney, 1947). However, the distributions for the LTS, the surface temperature and the MCAO index are significantly different. In particular, in the region of positive sensitivity, the LTS tends towards higher values, whereas the surface temperature and the MCAO index are both lower. This influence of stability on the LWP

sensitivity is consistent with previous studies (Chen et al., 2014); higher stability conditions inhibit mixing between the cloud layer with the dry above-cloud layer, and prevents the depletion of LWP due to the evaporation-entrainment mechanism.

The r^2 values of the correlation between the sensitivity and mean of meteorological variables for each pixel over the six years of data were calculated. The r^2 is higher for the LTS (0.39) than for the MCAO index (0.26) and surface temperature (0.32); the associated scatter plots are shown in Figure 4.4. This indicates that the LTS explains a greater fraction of the variance in the sensitivity than the other meteorological variables. Due to its better performance as an explanatory variable, LTS is used in the remainder of this study as a proxy for the importance of stability and surface forcing on cloud formation.

This association between positive sensitivities and higher LTS values explains why the strongest positive signal was observed during JJA in Figure 4.2. During the summer, the ocean temperature is constrained by melting sea ice, while the atmosphere is warmed by an increase in solar radiation, creating a weak ocean-air temperature gradient (Kay and Gettelman, 2009; Persson, 2012; Morrison et al., 2019). This results in warm air residing above a relatively cold ocean, producing high LTS conditions. Conversely, in spring and autumn, lower stability conditions are more common. Due to its large thermal heat capacity, the ocean remains warm relative to the atmosphere in seasons with low insolation, generating a larger ocean-air temperature gradient and unstable conditions (Kay and Gettelman, 2009; Morrison et al., 2019; Huang et al., 2019).

From Figure 4.3, it appears that the specific humidity does not strongly drive the LWP response. However, previous work has shown that specific humidity often strongly influences the N_d -LWP relationship, with more weakly negative responses under

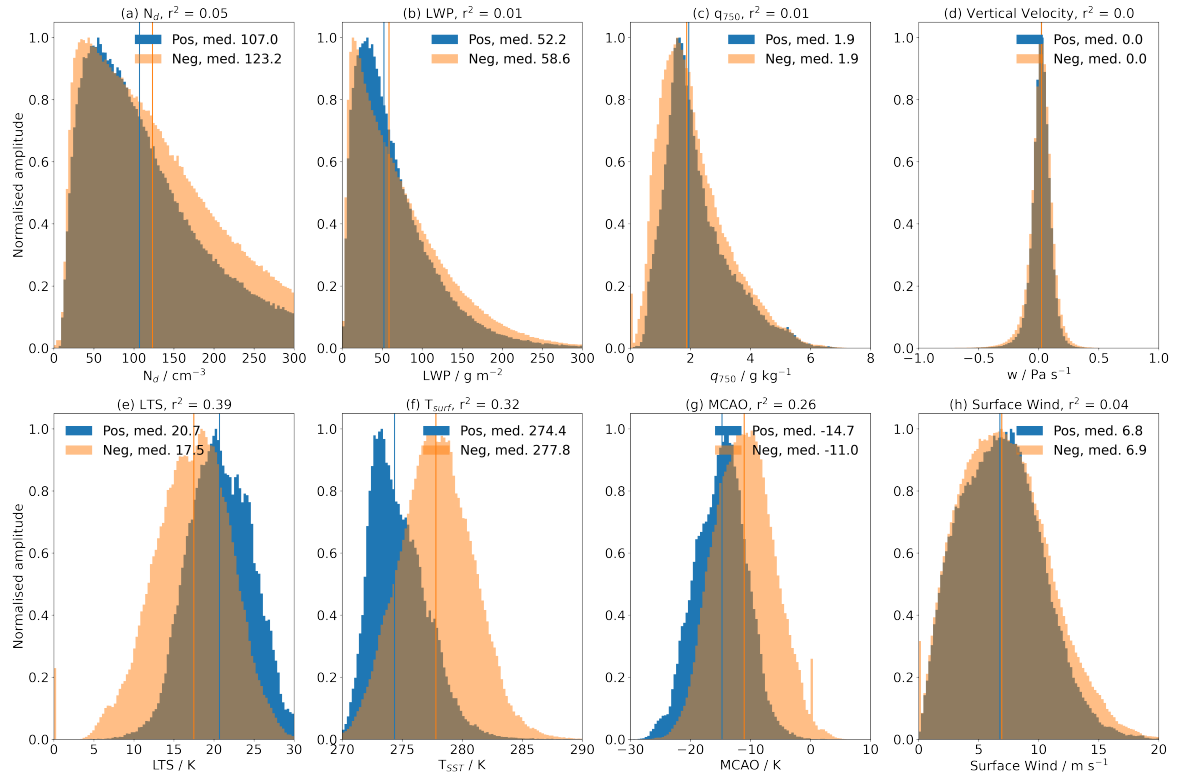


Figure 4.3: Normalised histograms showing the distributions of (a) N_d , (b) LWP, (c) q_{750} , (d) vertical velocity, (e) LTS, (f) T_{surf} , (g) MCAO and (g) surface wind for the regions of positive and negative sensitivity shown in Figure 4.2 (a). The blue and orange vertical lines show the medians for the positive and negative regions, respectively. The r^2 values relate to the correlation between the sensitivity and mean of meteorological variables for each pixel over the six years of data.

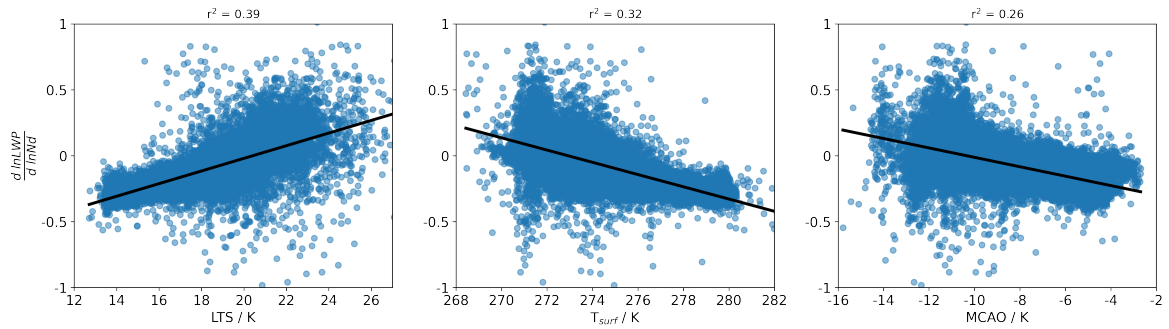


Figure 4.4: Scatterplots of the N_d -LWP sensitivity for each pixel in Figure 1 (a) plotted against the mean value of (a) LTS (b) T_{surf} and (c) MCAO index for that variable over the 6 years considered. The line of best fit is shown in black and the r^2 value is shown above each plot.

higher humidity conditions (Chen et al., 2014; Toll et al., 2019) due to a suppressed evaporation-entrainment mechanism. To investigate this further, the data were partitioned into bins of specific humidity and LTS (Figure 4.5). The response to changes

in specific humidity is weak, with strongly negative sensitivities evident under both humid and dry conditions. The response to variations in LTS is greater, with a strong negative response at low LTS turning into a positive sensitivity in higher stability conditions. The strong dependence on stability supports the hypothesis that the LTS is the predominant control behind the differences between the regions of positive and negative sensitivity (Figure 4.3).

Although the overall response to humidity is weak, Figure 4.5 shows that its role is dependent upon the LTS conditions. The influence of humidity on the N_d -LWP relationship is small at low LTS; the sensitivity is consistently strongly negative across the humidity range. However, it becomes important at high LTS, where the response changes from negative to positive as humidity increases. These results are similar to previous work on Arctic clouds; in high stability environments, Coopman et al. (2016) found an increasingly positive response with q_{750} . Additionally, the sensitivity to aerosol increased with LTS when q_{750} was constrained between 2.0 and 4.0 g kg⁻¹.

The results presented so far have assumed a linear sensitivity of LWP to N_d ; however, assuming linearity means important characteristics of the relationship are not considered. For example, the response of LWP can be non-linear, with a dependence on the initial cloud state (Gryspeerdt et al., 2019a). Additionally, use of the linear sensitivity parameter does not consider the absolute values of the LWP and how these change in different meteorological regimes. To investigate these characteristics, joint probability histograms were generated by creating a 2D histogram of LWP and N_d and then normalising each column by the total N_d , such that each pixel in that column represented $P(\text{LWP}|N_d)$, or the probability of observing a particular LWP given a particular N_d . These diagrams also allow for exploration of where along the N_d spectrum the meteorological conditions considered in Figure 4.5 become important

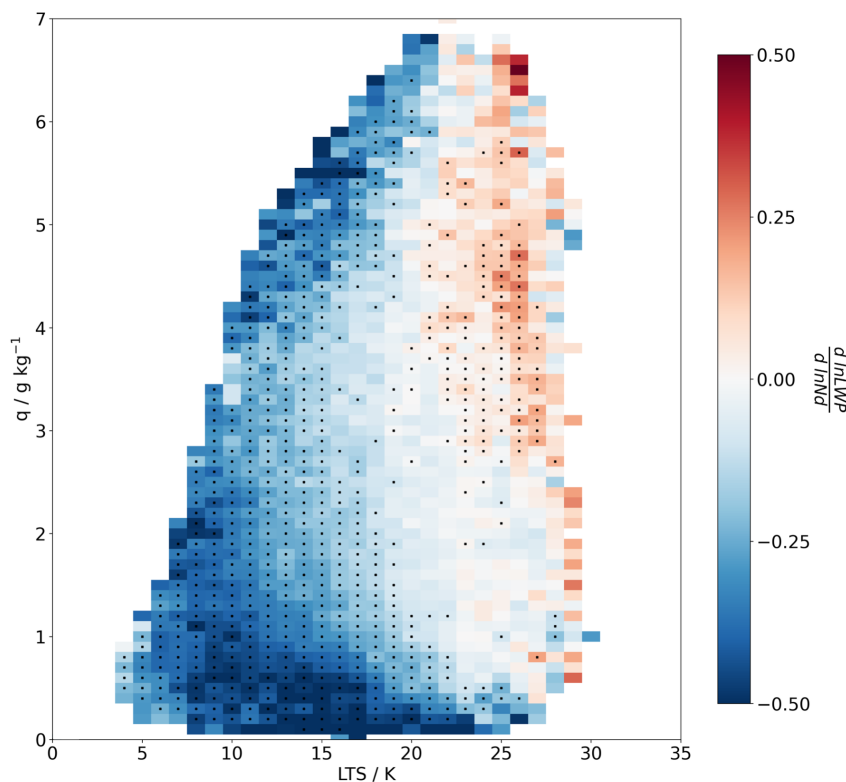


Figure 4.5: The linear N_d -LWP sensitivity plotted as a function of LTS and cloud-top humidity (q_{750}). Only bins which included over 100 valid retrievals were included in the analysis. The black dots indicate points for which the correlation is significant at a 95% confidence level.

to the N_d -LWP relationship.

Figure 4.6 shows the N_d -LWP joint probability histogram for four different environmental regimes, partitioned into high and low LTS and specific humidity bins. The differences between the normalised histograms are shown at the end of each row and column, and the N_d distributions for the regimes are shown beneath each histogram. The blue lines on the difference plots shows clouds with droplet effective radius of $15 \mu\text{m}$; clouds to the left of this line are assumed to be precipitating. The black lines on the joint probability histograms are the mean LWP for each N_d bin, with the shading showing the 95% confidence interval. These mean LWP lines are expanded in Figure 4.7 for clarity. Figure 4.7 also displays the seasonal breakdown of this relationship, showing dependence of the N_d -LWP on LTS and q_{750} is robust

across seasons, as would be expected from an effect driven by meteorology.

In high LTS conditions, Figures 4.6 (a) and (b) show evidence of three different responses of LWP to N_d , which are related to the precipitation behaviour as a function of r_e and the pixel aggregation scale. First, in clean states (low N_d ; up to 20 cm^{-3}), there are strong increases in LWP with N_d , as has been observed in previous work (Gryspeerd et al., 2019a); this is consistent with precipitation suppression. There is little difference in the high and low q_{750} environments in these low N_d conditions (Figure 4.6 (c)), suggesting that the precipitation suppression mechanism is not strongly reliant on specific humidity. The N_d distributions below the histograms show that these very low N_d conditions are relatively rare over the Arctic ocean and therefore have little bearing on the linear sensitivity.

In the second regime, between 50 cm^{-3} and $150 - 200 \text{ cm}^{-3}$, LWP still increases with increasing N_d , but the strength of the increase is not as large as for the clean cases (Fig 5 (a) and (b)). This is likely due to the precipitation suppression and evaporation-entrainment mechanisms competing to modulate LWP. Figure 4.8 shows that at these droplet number concentrations, there is still a significant proportion of the droplets which are sufficiently large ($r_e > 15 \mu \text{ m}$) for the precipitation suppression to augment LWP. However, most droplets are smaller than $15 \mu \text{ m}$, so the precipitation suppression mechanism is weaker than at lower N_d . In high LTS conditions, the evaporation-entrainment mechanism is suppressed enough that N_d increases up to $150 - 200 \text{ cm}^{-3}$ can still lead to increases in LWP. The similarity between the high and low humidity cases for this N_d range in Figure 4.6 (c) also suggests that this N_d -LWP relationship in this regime is not dominated by the evaporation-entrainment mechanism. From the N_d distributions, the majority of the data is in this regime and therefore it strongly influences the overall N_d -LWP sensitivities reported in Figures 4.2 and 4.5.

Finally, the LWP decreases with further increases in N_d in heavily polluted (above 200 cm^{-3}) environments. At these N_d , droplets are rarely larger than $15 \mu\text{m}$ (Figure 4.8), so the precipitation suppression mechanism can't generate further increases in cloud water. Therefore, the evaporation-entrainment mechanism becomes the dominant control and begins to reduce LWP. Figure 4.6 (c) shows that the LWP decreases are stronger in low humidity environments due to the enhanced evaporation of cloud droplets under these conditions. At high q_{750} , free tropospheric moisture can buffer against cloud water loss.

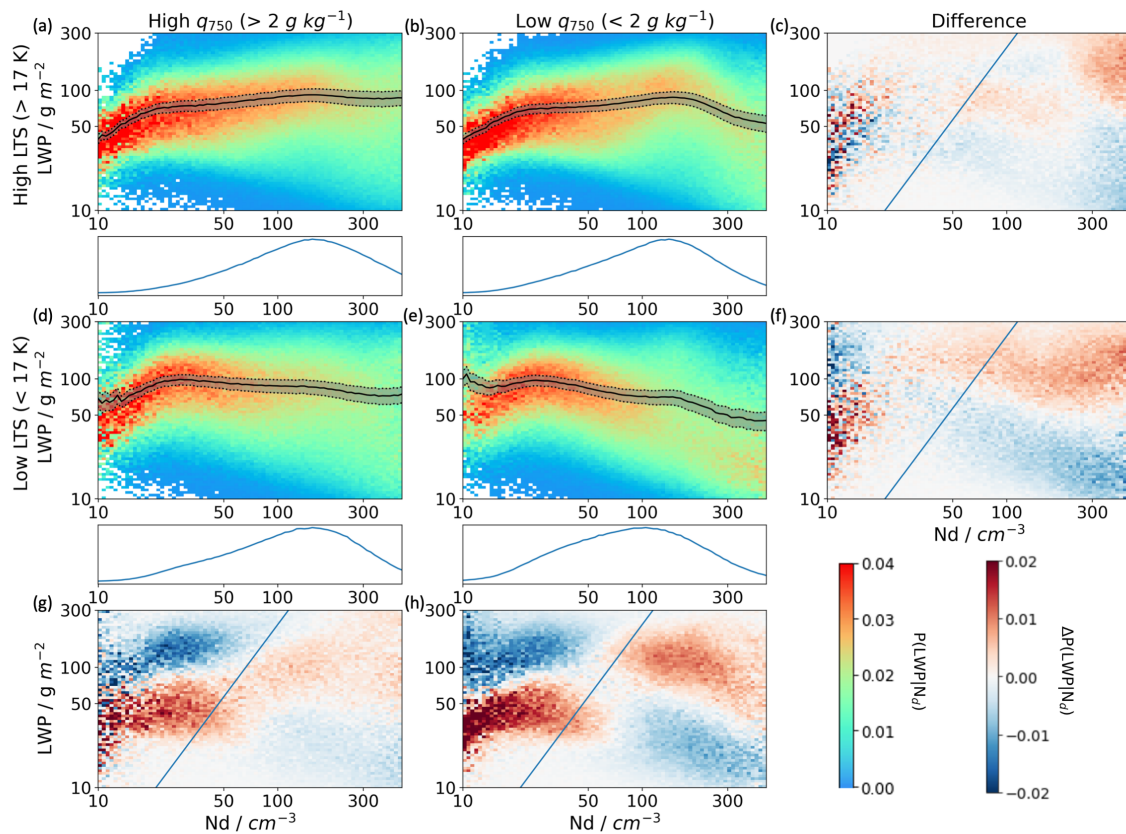


Figure 4.6: Joint probability histograms for the N_d -LWP divided into four meteorological regimes based on LTS and q . The difference plots are shown at the end of each row and column, with red over blue indicating higher LWP at higher humidity/LTS. The black lines and grey shading on the joint probability histograms represent the mean LWP value for each N_d bin and the 95% confidence interval, respectively. The blue lines on the difference plots indicate clouds with effective radius of $15 \mu\text{m}$, so clouds to the right of the line are expected to be non-precipitating.

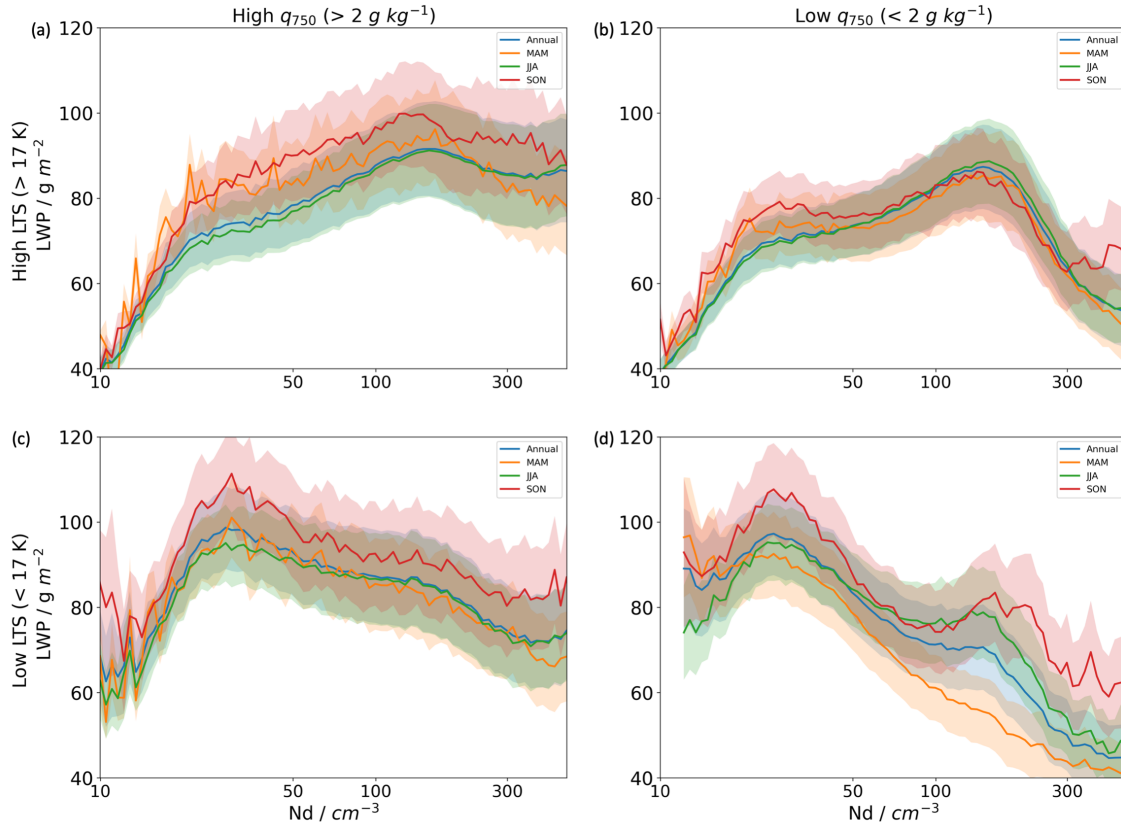


Figure 4.7: The mean LWP for each N_d bin and the 95% confidence interval for different meteorological regimes. The blue line is for all seasons and equivalent to the black lines in Figure 4.6, and the orange, green and red lines are for MAM, JJA and SON respectively.

Similarly to the stable regimes, the precipitation suppression mechanism is again evident for N_d up to $20 - 30 \text{ cm}^{-3}$ in low LTS environments. After this, the LWP decreases with increasing N_d , with stronger decreases seen in low q_{750} conditions (Figure 5 (c) and (d)). This is consistent with the evaporation-entrainment mechanism. Figure 4.6 (f) shows that the difference between the high and low humidity regimes manifests at a lower droplet number than in the high LTS regimes; in Figure 4.6 (c), clouds in high q_{750} environments do not have significantly greater LWP than clouds in low q_{750} environments until N_d reaches above 250 cm^{-3} (indicated by the darker red over blue region Figure 4.6 (c)). However, in Figure 4.6 (f), these larger LWP values in moister conditions occur earlier at around $40\text{-}50 \text{ cm}^{-3}$, with the disparity growing stronger with higher N_d . This importance of humidity at lower N_d at

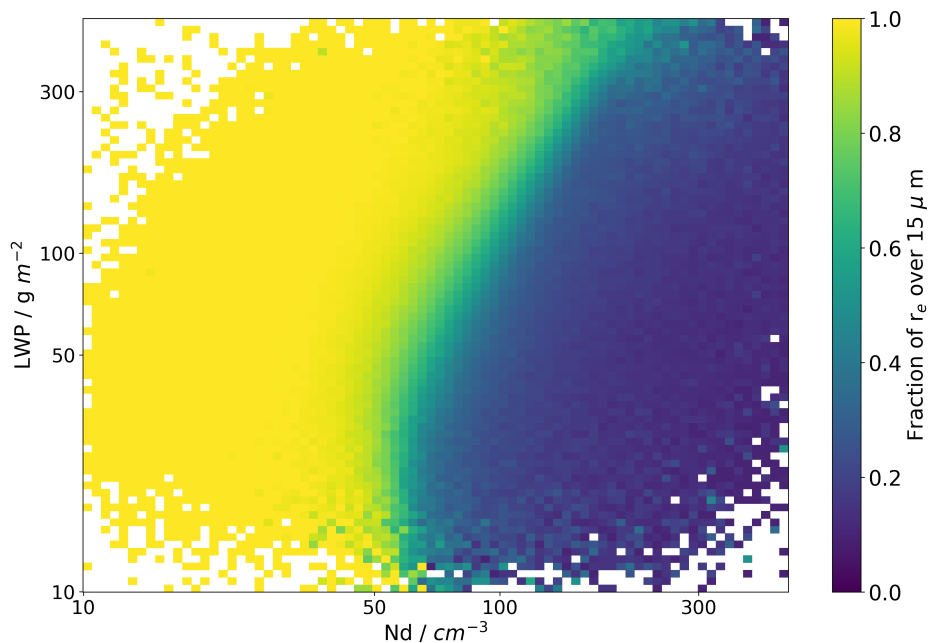


Figure 4.8: Fraction of 1km pixels with $r_e > 15 \mu\text{m}$ for a given 25km by 25 km grid, as a function of LWP and N_d . Droplets with $r_e > 15 \mu\text{m}$ are assumed to be precipitating. The threshold of $15 \mu\text{m}$ for collision-coalescence is taken following Rosenfeld and Gutman (1994).

low LTS may be due to increased turbulent mixing with the above-cloud layer. This would enhance the rate of droplet evaporation, thereby increasing the dependence of LWP on q_{750} .

When comparing across stability regimes, Figures 4.6 (g) and (h) show that when the N_d is below 50 cm^{-3} , low LTS conditions support clouds with a higher LWP. This may be due to a deepening of the boundary layer under unstable conditions, allowing clouds to grow deep enough to precipitate, such that an increase in aerosol allows for more LWP to be retained in the cloud through the precipitation suppression mechanism. Additionally, low LTS facilitates the vertical transport of moisture, thereby promoting cloud formation (Kay et al., 2016). This behaviour is similar in both high and low q_{750} environments, suggesting that humidity plays a smaller role at low N_d .

However, as N_d increases above $50\text{-}100\text{ cm}^{-3}$, Figures 4.6 (g) and (h) show that higher LTS conditions support higher LWP clouds than low LTS conditions. Stronger mixing with subsaturated air in low LTS environments may enhance droplet evaporation relative to stable environments. Drier cloud tops would strengthen this effect, resulting in the disparity in cloud LWP for high and low LTS being stronger in low q_{750} environments (Figure 4.6 (h)).

4.3.3 Radiative impacts

The potential radiative impact of the LWP adjustments relative to the albedo enhancements from more numerous, smaller droplets (the Twomey effect) can be estimated from :

$$\frac{d \ln \alpha}{d \ln N_d} = \frac{1 - \alpha}{3} \left(1 + \frac{5}{2} \frac{d \ln LWP}{d \ln N_d} \right) \quad (4.4)$$

In which α is the cloud albedo (following Platnick and Twomey, 1994). An estimate of the N_d -LWP sensitivity can be generated by using the present-day N_d distribution and the joint probability histograms in Figure 4.6 to generate a LWP distribution. The present day N_d distribution is then adjusted such that the mean increases by 10%, to represent an arbitrary hypothetical increase in aerosol concentration in a future Arctic. A new LWP distribution is calculated using this adjusted N_d distribution and the joint probability histograms. The change in the mean LWP and mean N_d between the two sets of distributions is calculated to estimate the sensitivity. Using this method, in high LTS conditions (ignoring the effects of humidity), LWP adjustments imply an 8% enhancement of the Twomey effect (slope of +0.03), and under low LTS conditions, an offset of 43% (slope of -0.17). The sign of the sensitivity is insensitive to the size of the perturbation for moderately sized adjustments to the N_d

distribution.

4.4 Discussion

This chapter investigated the factors which influence the N_d -LWP relationship in Arctic clouds. It was found that LTS is a dominant control on the LWP response, with increases in LWP possible in high stability conditions. Specific humidity only appears to influence the relationship in polluted or high LTS conditions, while the N_d state exerts little control on the LWP response. However, despite careful filtering to remove instances in which the data are prone to errors, some uncertainties in the results remain.

As LWP and N_d are both calculated using the τ_c and r_e retrievals, errors in these properties may result in significant correlated errors in the LWP and N_d MODIS retrievals. Gryspeerdt et al. (2019a) show that errors in τ_c would generate a positive bias, whereas errors in r_e would create negative bias in the magnitude of the relationship. However, when this analysis was repeated using the AMSR-E LWP (Figures 1 (b), 4.9 and 4.10), the LWP response to N_d across the different meteorological regimes was consistent with the results using the MODIS LWP. AMSR-E is an independent dataset and therefore not affected by the correlated errors in τ_c and r_e . Therefore, while correlated errors may affect the LWP- N_d relationship, they do not dominate the results and the observed relationships are not just retrieval artefacts. The joint probability histograms generated using the AMSR-E LWP and MODIS N_d (Figure 4.9) were also used to repeat the calculations in Section 4.3.3 to understand the effects of correlated errors on the estimate of the radiative impacts. In high LTS conditions, the AMSR-E LWP adjustments imply a 13% enhancement of the Twomey effect (slope of +0.05), and under low LTS conditions, an offset of 10% (slope - 0.04). The shift in the sign of the relationship with LTS is still evident, with changes

in unstable conditions unlikely to be positive and enhancements in stable conditions unlikely to be very large. However, due to uncertainties in AMSR-E retrievals, including underestimating LWP at values above 180 g m^{-2} (Section 3.5.2), the exact value of the enhancement or offset is unclear. A full understanding of the correlated errors is beyond the scope of this work.

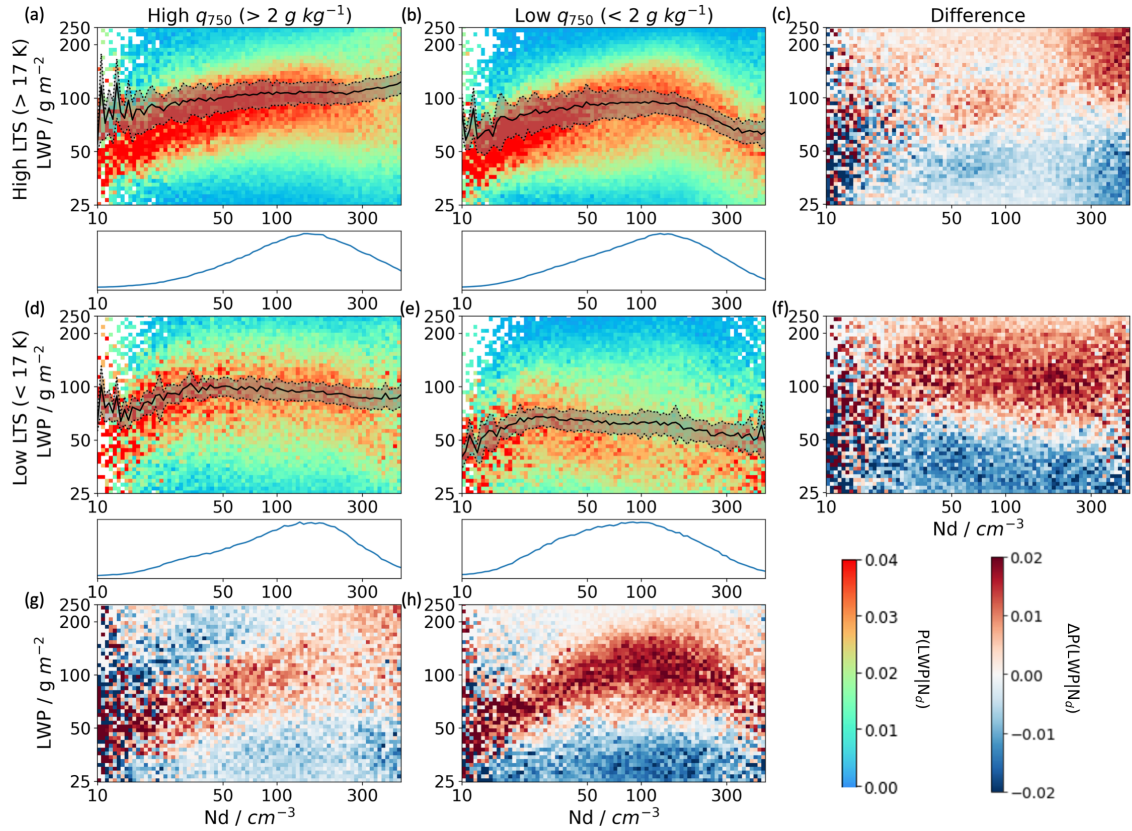


Figure 4.9: Joint probability histograms for the AMSR-E LWP-MODIS N_d divided into four meteorological regimes based on LTS and q . The difference plots are shown at the end of each row and column, with red over blue indicating higher LWP at higher humidity/LTS. The black lines and grey shading on the joint probability histograms represent the mean LWP value for each N_d bin and the 95% confidence interval, respectively.

Despite ERA5 performing better than other reanalysis datasets when compared to in situ observations of temperature and humidity (Graham et al., 2019b; Renfrew et al., 2021), particularly for ice-free areas of the Arctic, the meteorological conditions in the Arctic are still poorly constrained (Section 2.2). Therefore, use of the

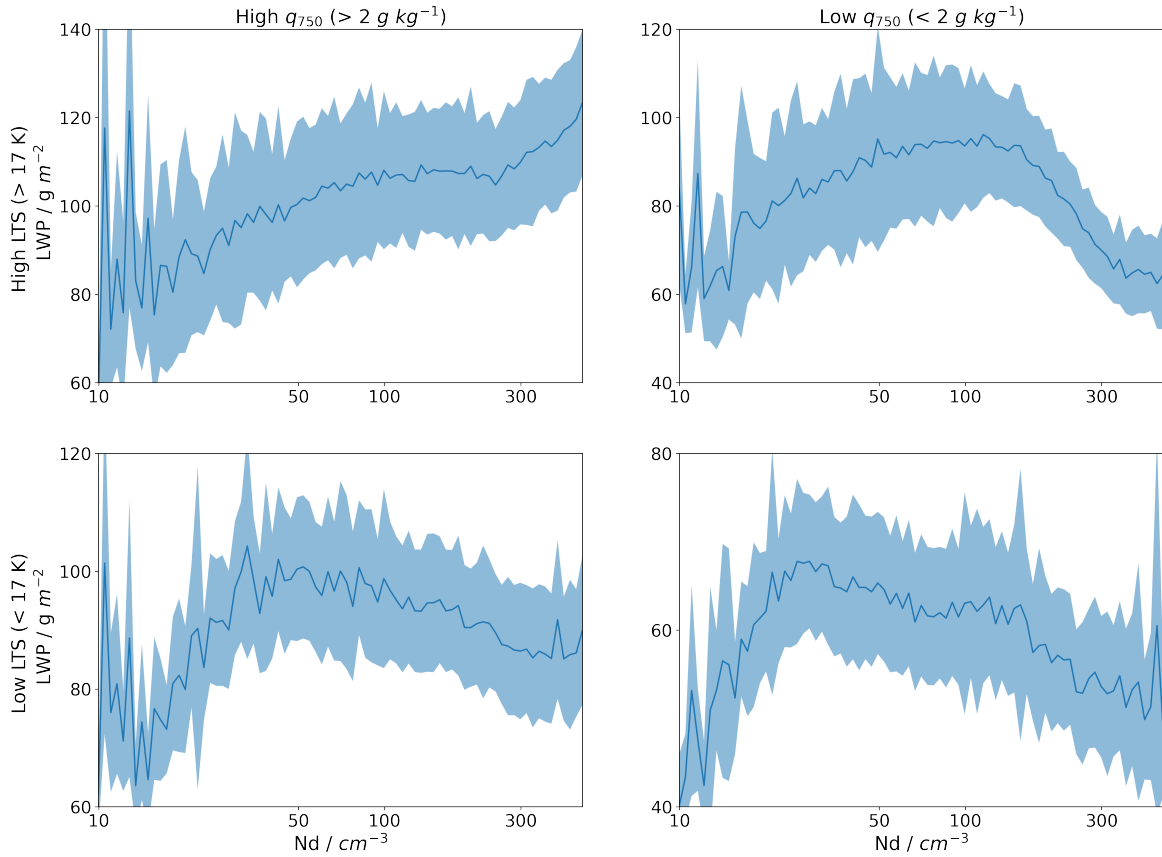


Figure 4.10: The mean AMSR-E LWP for each MODIS N_d bin and the 95% confidence interval for different meteorological regimes. The blue line is for all seasons and equivalent to the black lines in Figure 4.9.

reanalysis data may introduce additional uncertainties into the results.

As the Arctic warms, the LTS is projected to decrease (Boeke et al., 2021). Figure 4.6 shows that in lower LTS environments, LWP typically decreases with N_d , which weakens shortwave cooling effect of the clouds on the surface. Assuming moderate increases in N_d , the LWP adjustments shift from amplifying the Twomey effect by 8% in a stable environment to a 43% reduction in an unstable one. This change in the sign of the N_d -LWP relationship with warming amounts to a temperature-dependent indirect effect; in a warmer Arctic with a more unstable boundary layer, LWP adjustments may shift from enhancing the cooling effect to offsetting it.

Equally, these results also demonstrate an aerosol-dependent cloud feedback, as the LWP response to changes in LTS is different in clean and polluted environments. In more polluted environments, LWP decreases more strongly as LTS decreases (Figure 4.6 (g) - (h)). This weakens the negative cloud feedback, shifting to a positive effect. This influence of aerosols on the cloud feedback is key as industrialisation and the creation of new trans-Arctic shipping lanes are projected to be developed as the Arctic heats and sea ice retreats, introducing a large new source of anthropogenic aerosols (Peters et al., 2011; Schmale et al., 2018). However, there is considerable uncertainty about the effects of these new aerosol emissions, particularly as remote sources of aerosols are project to decrease as countries introduce more stringent air quality guidelines (Schmale et al., 2022b).

In stable conditions, the N_d -LWP relationship is positive, with decreases in LWP only seen in heavily polluted environments (Figure 4.6 (a) and (b)). Additionally, in relatively clean conditions, clouds in low LTS environments have higher LWP than clouds in stable environments (Figure 4.6 (g) and (h)). Therefore, individually, the increases in aerosol or decreases in LTS projected for the Arctic may act to strengthen the cloud shortwave effect. However, working together in a warmer Arctic they may produce clouds with lower water paths, leading to a weaker negative cloud feedback in a more polluted environment.

The results presented here are only for liquid clouds over ocean; more work is required using different datasets to see if they hold in ice-covered regions or for mixed-phase clouds. Nevertheless, these findings that the aerosol-cloud interactions change with warming and that the LWP-LTS relationship depends on the aerosol loading may have significant implications for the surface energy budget in a rapidly changing Arctic. For example, potential decreases in cloud LWP suggested by this study could have significant consequences for Arctic sea ice extent. Thinner clouds have

a lower albedo, and therefore a lower shortwave cooling effect at the surface. In summer months, when surface albedo is low due to the presence of open ocean, this leads to an increase in solar radiation being absorbed by the surface. Previous work has found a negative correlation between the amount of radiation absorbed by the surface in summer, which is in part controlled by cloud LWP, and sea ice extent later in the year (Choi et al., 2014; Huang et al., 2017). The effects of these LWP changes to the longwave effect, which dominates in non-summer months, are expected to be weaker, as Arctic clouds in non-summer months typically have LWP greater than 30 g m^{-2} , and therefore act as black-body radiators (Shupe and Intrieri, 2004; Huang et al., 2017, 2019). As a result, changes in cloud fraction control the longwave downwelling radiation in the Arctic.

4.5 Conclusions

Previous studies have found a strong sensitivity of Arctic cloud properties and aerosols (Garrett et al., 2004; Coopman et al., 2018a). However, these works were either of a limited spatial extent or considered the average response of cloud properties across the Arctic, and therefore did not observe the spatial heterogeneity in cloud response. This work considered the regional variation in the LWP response to N_d in liquid clouds, documenting a positive sensitivity at higher latitudes. Positive relationships have previously been observed under some conditions, but not at the strength found in this work (Han et al., 2002; Chen et al., 2014; Toll et al., 2019; Gryspeerdt et al., 2019a). However, the response is typically negative across the globe. The signal was most strongly observed during the summer months (Figure 4.2). Comparison of cloud and meteorological properties of the regions displaying positive and negative sensitivity indicates that stability, in particular LTS, is a significant driving force for the difference in behaviour (Figure 4.3).

There is only a weak response to cloud-top specific humidity, but the variation with LTS was much greater (Figure 4.5). Under moist, stable conditions, the LWP increases with N_d , as seen with subtropical clouds (e.g. Chen et al., 2014). Even when considering cases with lower humidity, increases in LWP with N_d are supported up until high N_d , at which point the humidity is insufficient to offset the moisture lost to droplet evaporation (Figure 4.6). The frequency of these high LTS conditions at high latitudes during the summer months explains the seasonal pattern in the sensitivity in Figure 4.2.

This work found evidence for a temperature-dependent aerosol indirect effect through the change in sign of the N_d -LWP relationship with LTS and an aerosol-dependent cloud feedback as the LWP response to LTS, and therefore surface temperature, changes with N_d . Unstable conditions generate higher LWP values than stable conditions for low N_d , potentially due to precipitation suppression (Figure 4.6). Therefore, in a future, lower-LTS environment, clouds have a stronger shortwave cooling effect. However, the response to aerosol in a warmer Arctic is different; interactions with aerosols would produce lower LWP clouds, thereby reducing the aerosol cooling effect. Using the Twomey relation (Equation 4.3.3) and simple assumptions about the future environment, an increase in N_d and move to low LTS conditions implies that the N_d -LWP relationship shifts from slightly enhancing the Twomey effect to offsetting it by 43%.

These findings on the dependence of the N_d -LWP relationship on the LTS and mean N_d state have important consequences for cloud feedbacks in a warmer, more polluted Arctic. The combined effect of increasing aerosol concentrations in conjunction with increases in surface temperature and decreases in the LTS may ultimately lead to thinner, lower-LWP clouds, with a reduced cooling potential.

However, an important limitation of this work is its reliance on deriving statistical relationships from snapshot satellite imagery. While this method has been widely used in previous work investigating aerosol-cloud interactions (e.g., Chen et al., 2014; Michibata et al., 2016; Gryspeerd et al., 2019a), the effects of aerosols on cloud properties occurs over a range of timescales, from the near-instantaneous Twomey effect to the longer LWP adjustments. Therefore, the temporal averaging performed here may mask some of the complexity and may lead to erroneous inferences about the nature of the N_d -LWP relationship.

Considering the LWP response with time may help towards establishing the causal relationship between aerosol perturbations and changes in cloud properties. Constraining causality is crucial for developing a mechanistic understanding of the effects of confounding variables, such as meteorological conditions. Additionally, gaining an understanding of the timescales for cloud property responses is useful for model development, as models often struggle to simulate microphysical processes. Model evaluation often comprises comparing mean states with satellite imagery, without necessarily considering how these dynamic processes are represented. Building a time-resolved picture of cloud evolution may allow for a better diagnosis of the persistent problems in how models represent cloud properties. Investigating the evolution of Arctic cloud properties with time will be the subject of subsequent chapters of this thesis.

Chapter 5

Investigating the development of clouds within marine cold air outbreaks

Key Points

- Marine cold air outbreaks (MCAOs) are associated with characteristic evolution in cloud properties, which transition from high-coverage, stratiform clouds to broken, low-coverage cumuliform cloud fields.
- While the initial strength of MCAOs influences cloud property development, the timing of the cloud transition is not significantly altered.
- Higher initial aerosol is linked with a delay in the cloud transition by 5-7 hours and a slower decline in cloud fraction.
- Precipitation appears to be key to triggering the transition, as the timing of LWP breakdown is nearly coincident with the point at which the mean droplet effective radius reaches precipitation threshold.
- The cloud fraction is the most dominant control on cloud albedo.

The contents of this chapter are based on Murray-Watson, R. J., Gryspeerdt, E., and Goren, T.: Investigating the development of clouds within marine cold-air outbreaks, Atmos. Chem. Phys., 23, 9365–9383, <https://doi.org/10.5194/acp-23-9365-2023>, 2023. The analysis and text are my own work, with comments from my co-authors.

5.1 Introduction

As discussed previously, achieving a better understanding of cloud processes is crucial to improving their representation in models. While Chapter 4 shows that the shortwave radiative effect of liquid clouds may respond to changes in both surface temperature and aerosols, the approach taken does not account for cloud development, which may also be expected to change with these factors. Recently, there has been a drive to use existing satellite data in novel ways to move towards a pseudo-Lagrangian framework for studying cloud development (e.g., Christensen et al., 2020; Gryspeerdt et al., 2021; Horner and Gryspeerdt, 2022). These and other works illustrate the importance of defining a suitable baseline, such that the signal arising from changes in cloud properties caused by aerosols is not masked by variability or variation in the initial conditions (Gryspeerdt et al., 2022a). In the Arctic, particular mesoscale phenomena associated with cloud formation offer a unique opportunity to investigate cloud development from a distinct starting point: marine cold air outbreaks (MCAOs).

MCAOs form due to polar or cold continental air moving over a relatively warm ocean surface. The vertical temperature gradient generates intense turbulent heat and moisture surface fluxes, promoting cloud formation (Brümmer, 1996; Fletcher et al., 2016a; Papritz and Spengler, 2017). Outbreak events can last several days and reach scales of up to 1000 km (Fletcher et al., 2016a; Kolstad, 2017). MCAO clouds can be characterised in satellite imagery as cloud ‘streets’ (stratocumulus decks)

moving to a broken, cumuliform cloud field downwind (Brümmer, 1999; Pithan et al., 2018), as seen in Figure 5.1. These events offer polar analogues to the subtropical stratocumulus-to-cumulus transitions, which have previously been used to study cloud life cycles (e.g., Sandu and Stevens, 2011; Christensen et al., 2020). MCAOs, with their distinct development and clear baseline, offer a ‘natural laboratory’ for studying the impact of factors like initial aerosol concentration on cloud formation. These high-latitude events also offer the unique advantage of being captured by frequent MODIS overpasses, up to 16 times a day, allowing for a detailed, time-resolved analysis of cloud development.

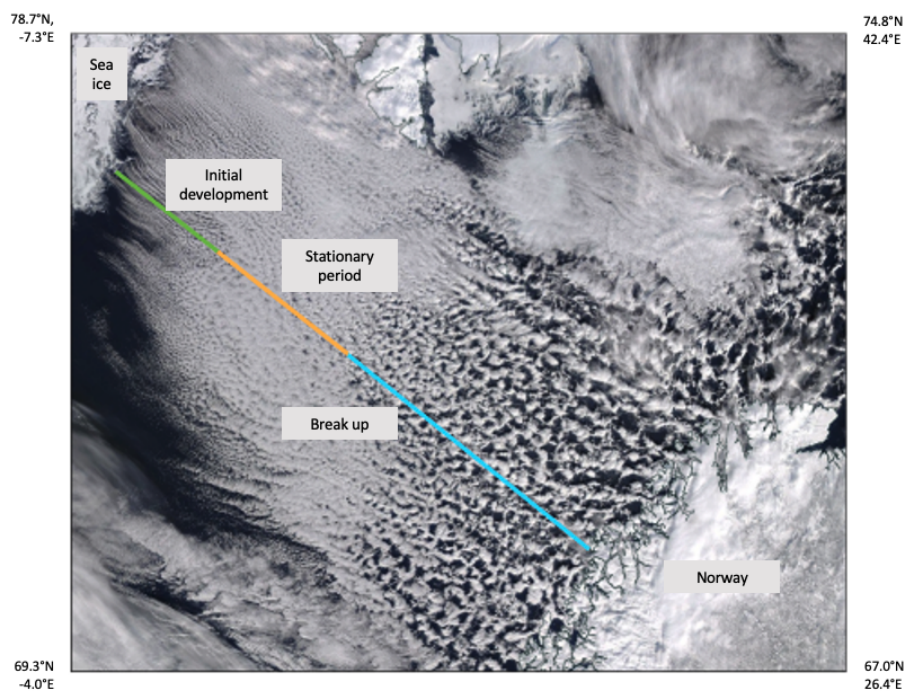


Figure 5.1: Cloud development in a marine cold air outbreak on 21st March 2021, moving from the ice edge to the Norwegian coastline in the bottom right. Source: NASA Worldview, Corrected Reflectance (True Color) from the MODIS instrument on Terra.

Previous work on the subtropical stratocumulus-to-cumulus transition have identified several causes as their driving force, such as an increasing sea surface temperature gradient and precipitation mediated by aerosol and ice-production processes

(Sandu and Stevens, 2011; Christensen et al., 2020; Sarkar et al., 2020). As air is advected over the relatively warm ocean surface, the strength of the turbulent surface fluxes deepens the boundary layer, eventually causing the cloud layer to be decoupled from the surface (Bretherton and Wyant, 1997; Sandu and Stevens, 2011). As the ocean surface is cut off as a source of moisture and aerosols, the stratocumulus decks eventually dissipate. However, the below-cloud layer continues to be warmed and moistened by the ocean surface, forming cumuliform clouds in the lower boundary layer. Although primarily studied for subtropical clouds, McCoy et al. (2017) found that boundary layer instability and surface forcing were key drivers for developing open-cell cloud morphologies in MCAO.

Precipitation can also facilitate the break-up of cloud fields; the evaporation of precipitation below the cloud cools and moistens the air of the sub-cloud layer (Stevens et al., 1998; Yamaguchi et al., 2017; Tornow et al., 2021). This cooling creates instability near the surface, which, in conjunction with the moistening effect, is favourable to the formation of cumuliform clouds (Stevens et al., 1998). All else being equal, earlier precipitation would cause a more rapid transition. Precipitation also reduces the lifetime of the stratiform cloud layer by the removal of water (Abel et al., 2017; Lloyd et al., 2018). Frozen precipitation, in particular, has been identified as key to breaking up the cloud field in MCAO by accelerating the removal of cloud water through aerosol scavenging and mechanisms such as riming (Tornow et al., 2021) or secondary ice production (Abel et al., 2017; Karalis et al., 2022). Through modelling an MCAO north of the United Kingdom, Abel et al. (2017) found that precipitation, as opposed to the sea surface temperature gradient or entrainment drying of the cloud, was the key driver of cloud breakup.

Aerosols can strongly influence the onset of precipitation. A higher aerosol load generally leads to smaller liquid cloud droplets, which coalesce into precipitation-sized

droplets more slowly, potentially delaying the transition (Albrecht, 1989; Yamaguchi et al., 2017; Goren et al., 2019). Once rainfall begins, often through the build-up of cloud water, the falling precipitation removes boundary layer aerosols, creating a negative aerosol gradient over the outbreak (Abel et al., 2017; Lloyd et al., 2018; Dadashazar et al., 2021). This aerosol scavenging creates a positive feedback loop, as fewer aerosols mean a reduced number of sites on which new droplets can form, causing droplets to grow sufficiently large to precipitate (Jing and Suzuki, 2018). Modelling studies suggest that higher initial CCN concentrations delay the formation of precipitation in MCAO (Tornow et al., 2021). However, INPs can potentially accelerate the transition through droplet riming and enhancing precipitation (Abel et al., 2017; Tornow et al., 2021).

Global and high resolution models struggle to simulate evolution and properties of the low-level liquid and mixed-phase clouds often found in these outbreaks (Morrison et al., 2012; Field et al., 2014; Bodas-Salcedo et al., 2016; Abel et al., 2017; Field et al., 2017). Even higher-resolution numerical weather prediction models struggle due to the clouds falling into a convective ‘grey zone’ where the necessary dynamics cannot be parameterized (Field et al., 2017). The persistent negative biases in the shortwave reflectivity of Southern Ocean clouds in GCMs have been attributed to the representation of supercooled liquid (Cesana et al., 2022). The radiative properties of these clouds are of particular interest, given their prevalence and the changing climate in the region (Chapter 1.1).

As the processes which affect the MCAO cloud evolution are fundamentally time-dependent, knowledge of these process rates is essential for improving their representation in climate models (Pithan et al., 2018). Previous studies have used models (e.g Tornow et al., 2021), in situ or airborne measurements (Hartmann et al., 1997; Young et al., 2016; Abel et al., 2017; Lloyd et al., 2018; Ruiz-Donoso et al.,

2020; Geerts et al., 2022) and satellites (Wu and Ovchinnikov, 2022) to investigate the factors influencing cloud property development during the course of an outbreak. However, these were typically based on a relatively small set of examples of MCAO events. Geostationary satellites have been used to study clouds along the stratocumulus-to-cumulus transition following a large collection of Lagrangian trajectories in the subtropics (e.g. Christensen et al., 2020), but they do not cover high latitudes well.

Polar-orbiting satellites can provide several consecutive images for a location at high latitudes, providing a unique opportunity to characterise the temporal development of clouds. To study this evolution, I generate composite trajectories of air parcels moving off the ice edge using reanalysis wind fields. These trajectories are co-located with reanalysis data and data from several satellite instruments to investigate the factors which influence liquid cloud properties during the course of the cold air outbreak. In particular, I examine the controls on cloud properties that most strongly determine cloud albedo, namely cloud fraction and liquid water path (Loeb et al., 2007; Engström et al., 2015). The development of these cloud properties is then linked to changes in the top-of-atmosphere (TOA) albedo to estimate how these different environmental conditions affect the potential cloud radiative forcing.

5.2 Materials and Methods

5.2.1 Data

MODIS Level 2 Collection 6.1 cloud data (MYD06_L2; Platnick et al., 2017) were filtered and processed in a similar manner to Chapter 4. The differences in processing steps are outlined here. The period of study was between 2008 and 2014, inclusive. The area of interest is limited to the North Atlantic and Kara Sea region to avoid bias

towards shorter trajectories (Section 5.2.2).

It is possible that clouds embedded in the outbreaks are obscured by higher-level overlying clouds; to limit this effect, L2 pixels with cloud top heights above 500 hPa were eliminated; Fletcher et al. (2016b) showed that clouds within outbreaks are typically lower than this.

The LWP and N_d were calculated using Equations 2.2 (Wood and Hartmann, 2006) and 2.1 (Szczodrak et al., 2001; Quaas et al., 2006), with the same parameter values as Chapter 4.

Although ice is often present in MCAO clouds (Fletcher et al., 2016b), the factors which influence ice-phase processes and phase transitions are challenging to study using passive satellite data. The development of ice in these clouds will be considered further in Chapter 6. The focus here is on the development of liquid-dominated MCAO clouds. Grid cells (25 x 25 km) in which the MODIS sensor detect a non-zero ice fraction are removed from the analysis. Although the MODIS optical property phase algorithm typically performs well in comparison to active sensors (Marchant et al., 2016), it is still unlikely that this filtering entirely removes ice clouds from the dataset. Additionally, only pixels with a cloud top temperature above 263 K are included; in situ measurements in the Arctic have shown that these clouds typically have very high liquid water fractions (de Boer et al., 2009). This filtering to remove ice reduces the dataset to 31% of its original size.

DARDAR (Delanoë and Hogan, 2010; Ceccaldi et al., 2013) is used to analyse the efficacy of these filters to restrict the analysis to liquid-dominated clouds. Due to known issues with surface clutter affecting DARDAR retrievals, cases with cloud top heights below 720 m are not included. Figure 5.2 shows the DARDAR retrieved

cloud top phase fractions as a function of MODIS temperature for the set of MODIS pixels filtered to remove ice.

The phase fraction is calculated as described in Section 2.3. Figure 5.2 indicates that with this filtering, the supercooled phase fraction starts at 73% at 263 K and increases with temperature. Although DARDAR shows a non-negligible proportion of ice-containing phases, the real amount of ice present in these clouds is uncertain, as it would take a relatively small amount of ice to register a radar return and cause the retrieval to be classified as ‘mixed’ phase (Bühl et al., 2013). Restricting this study to pixels where DARDAR only registers supercooled liquid clouds would also reduce the volume of data available for analysis to less than 5% of the MODIS pixels co-located with the trajectories due to the DARDAR’s nadir-only sampling. As such, the method used here for filtering out ice-containing pixels is deemed sufficient for this work. The potential effects of biases introduced by ice undetected by MODIS will be discussed more in Section 5.4. Additionally, the presence of ice, mixed-phase and supercooled clouds above the freezing temperature may be due to either errors in the ECMWF temperature used to determine phase in DARDAR (Section 2.2) or biases in the MODIS cloud top temperature.

TOA albedo data were obtained from hourly Clouds and the Earth’s Radiant Energy System (CERES) SYN1deg L3 dataset (NASA/LARC/SD/ASDC, 2017). These data were regridded from the original $1 \times 1^\circ$ resolution to the same 25×25 km grid and projection as the MODIS data. Previous studies have shown that CERES TOA albedo measurements perform well over ocean (e.g. Sun et al., 2006; Kato et al., 2018), but are biased relative to in situ measurements if sea ice is present (e.g. Riihelä et al., 2017; Huang et al., 2022).

ERA5 reanalysis data was used for the meteorological variables (Hersbach et al.,

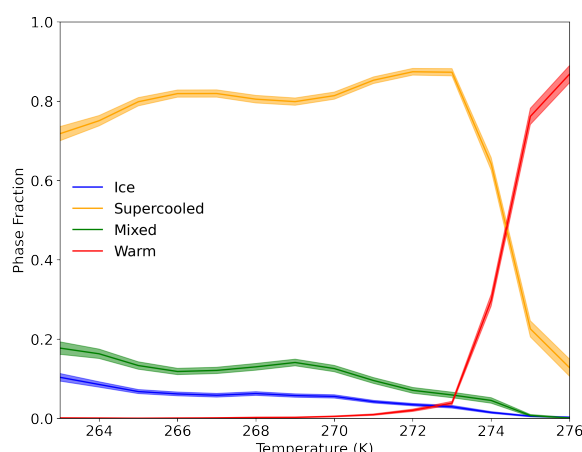


Figure 5.2: DARDAR cloud top phase fraction binned against MODIS cloud top temperature for clouds following the trajectories generated in this study. DARDAR phase flags ‘Ice’, ‘Spherical_or_2D_ice’ and ‘Highly_concentrated_ice’ are combined to calculate the ‘ice’ fraction. DARDAR flags ‘Supercooled’, ‘Liquid’ and ‘Supercooled_and_ice’ are considered to be ‘Supercooled’, ‘Warm’ and ‘Mixed’, respectively. Phase fraction is calculated by dividing the number of successful retrievals for a given phase flag by the total number of DARDAR retrievals for a 25 km by 25 km pixel. The shading represents the 95% confidence interval.

2020). The wind data at 1000 hPa was chosen to represent the boundary layer wind speed (following Gryspeerd et al., 2021), and the specific humidity at 800 hPa was taken to represent the moisture conditions above the cloud top (based on mean cloud top pressure in MCAO, Figure 5.6 (g)). The total AOD at 550 nm reanalysis data from the CAMS (Inness et al., 2019), was used as an indicative measure of aerosol conditions in the region.

The marine cold air outbreak index (M; Kolstad and Bracegirdle, 2008) is an important indicator of cloud formation and behaviour at high latitudes. It measures the stability of the boundary layer and is calculated as the difference between the potential temperature at 800 hPa and the sea surface temperature (Fletcher et al., 2016a). This metric is particularly suitable for the air moving off the ice edge as it highlights the difference in temperature of the relatively warm ocean with the cool overlying air masses. In the Northern Hemisphere, outbreak events are most common in the winter, followed by autumn and spring (Fletcher et al., 2016a). MCAOs are rela-

tively rare in the summer. As some cloud properties, such as r_e , can only be retrieved during sunlight hours, this analysis is restricted from March to October each year. Figure 5.3 (a) shows the mean MCAO index (M) within outbreaks (excluding times when $M \leq 0$) and the relative frequency of occurrence of outbreaks (Figure 5.3 (b)) for March to October 2008-2014.

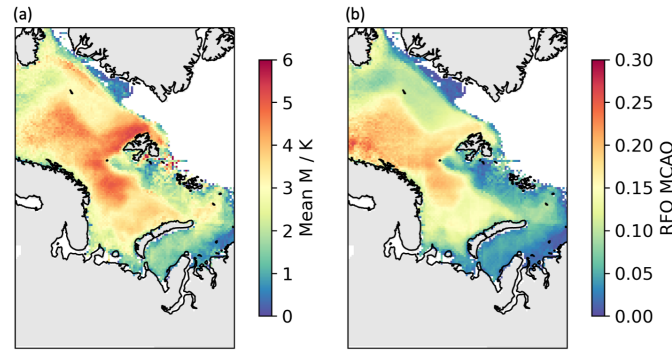


Figure 5.3: MCAO index (M) calculated from ERA5 data for March-October 2008-2014, filtered to only include MCAO events ($M > 0$) (a) and relative frequency of occurrence for outbreaks (b). Grey represents the land and white represents no data due to sea ice coverage.

5.2.2 Trajectory Generation

ERA5 reanalysis wind fields were used to create the pseudo-Lagrangian trajectories. Wind data at 1000 hPa were used for the advection, as this has previously been shown to follow low-level clouds successfully (following Gryspeerd et al., 2019b). Examples of the trajectories generated by this method are shown in Figure 5.4.

The advection procedure is adapted from Horner and Gryspeerd (2022), which focuses on trajectories starting from points of new convection. Here, a similar method is used, but the initial points are identified as cases where the air has moved from being over sea ice to being over open ocean between time steps of one hour. These pixels newly over the ocean are given a value of ‘time since ice’ (TSI) of one hour,

and all other pixels which did not move from ice to ocean remain at zero (Figure 5.4). Then, all pixels are advected forward again following the wind fields. For pixels previously identified as having moved off the ice edge in the first time step, one hour is added to their TSI. Pixels newly over ocean are again identified and given TSI values of one hour. This advection process repeats for every time step, with a value of one being added to all pixels on trajectories moving from the ice edge, a value of one hour assigned to pixels newly over the ocean, and zero assigned to all other pixels. In addition to the TSI value, each pixel has a date and time associated with the wind fields used to produce it; this is used to co-locate these pixels with the satellite data. If pixels move over land or back over sea ice, the trajectory is no longer followed. When two trajectories of two or more pixels converge, the smaller TSI is taken as the value for that pixel. The mean TSI for the region of interest is shown in Figure 5.5.

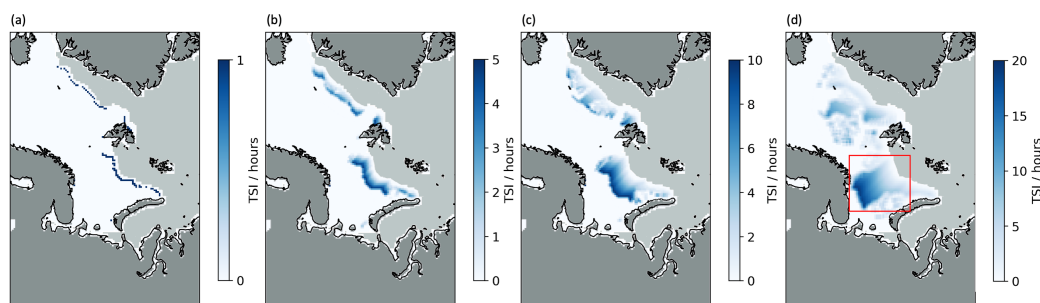


Figure 5.4: Snapshot of the ‘time since ice’ trajectory generation for (a) one (b) five (c) ten and (d) twenty hours since moving from the ice edge. Dark and light grey represent land and sea ice pixels, respectively. The scale varies between subplots. The red box in (d) indicates air moving off the ice edge that developed into a marine cold air outbreak. The data is from 1st April 2014.

A set of reverse trajectories of air parcels travelling over the ocean towards the ice edge were generated by running the above process in reverse; cases which move from being over open ocean to being over ice between time steps in the ‘forwards’ direction are identified, and then tracked backwards in time. These are useful to compare to clouds moving away from the ice edge as the effects of any retrieval

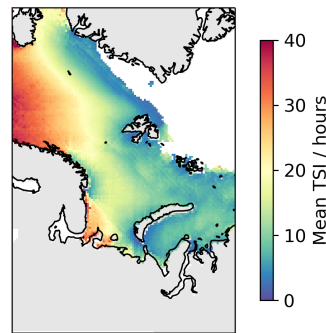


Figure 5.5: Map of the average time since ice for trajectories generated between March-October 2008-2014. Grey represents the land and white represents no data due to sea ice coverage.

biases, particularly close to the ice edge, are revealed. Additionally, the clouds along these trajectories are effectively blind to the ice edge but travel through generally similar environmental conditions over the ocean as those moving off of the ice edge. As a result, the changes in cloud properties resulting from moving off the ice edge can be highlighted. In these cases, ‘time towards ice’ (TTI) pixel values indicate the number of hours before the clouds reach the ice edge along these trajectories (i.e., a TTI of 30 hours means that, following this advection scheme, that pixel will reach the ice in 30 hours).

5.3 Results

5.3.1 Effects of moving off the ice edge

Figure 5.6 shows the development of cloud properties along three sets of trajectories: clouds approaching the ice edge (‘Towards’), clouds not embedded in cold air outbreaks (‘non-MCAO’) moving away from the ice and those within outbreaks (‘MCAO’). The MCAO and non-MCAO trajectories were partitioned based on M value early on in the trajectory ($M > 0$ within the first ten hours of leaving the ice edge).

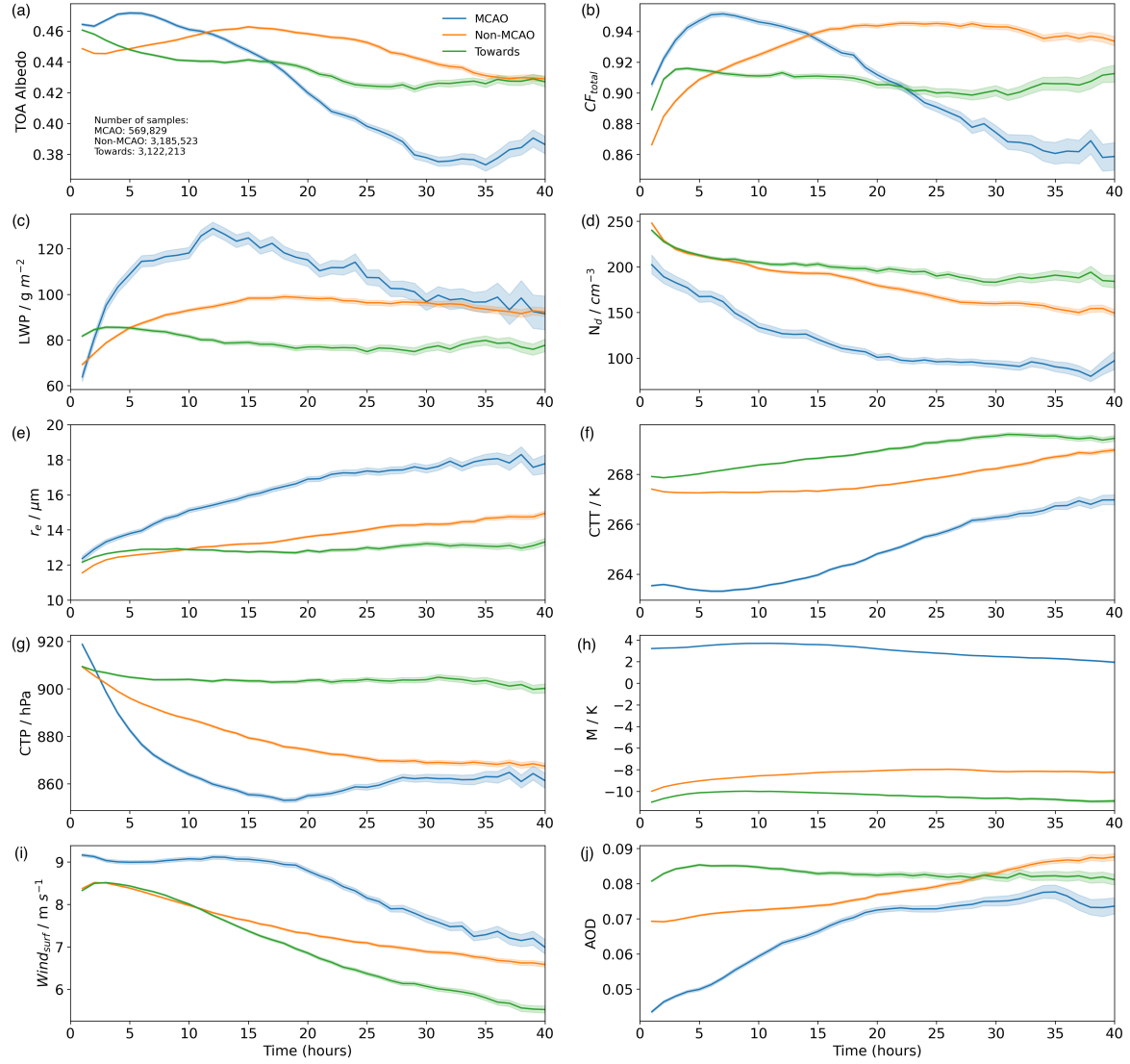


Figure 5.6: Cloud properties and environmental conditions along the MCAO (blue), non-MCAO (orange) and Towards (green) trajectories. CERES data was used for (a), and data for (b) - (f) were obtained from the MODIS instrument. Reanalysis data was used for Figures (g)-(j). CTT and CTP are the cloud top temperature and pressure, respectively. For all of the trajectories, a time of 0 hours represents the ice edge. For the MCAO and non-MCAO clouds, the time coordinate represents the ‘time since ice’, and their development moving from the ice edge proceeds from left to right in each subplot. For the Towards clouds, it is the ‘time towards ice’, or the number of hours until these air parcels reach the ice edge, and their development is instead read from right to left. The shading represents the 95% confidence interval (found using bootstrapping).

Cloud Fraction

Clouds moving towards the ice edge (the ‘Towards’ trajectories) maintain a consistently high cloud fraction (between 90% to 92%; Figure 5.6 (b)) over the observation period, with a sharp decrease near the ice edge. This decrease may be because of a retrieval bias, but as it does not appear to affect any of the other measured properties, I expect this to have negligible impact on the results. While high, these cloud fractions are typical for the region, particularly between spring and autumn in the Barents Sea, where most of the trajectories in this study were generated (Figure 5.6 (a); Kay et al., 2016). Non-MCAO clouds slowly increase to a peak of 94% at 22 hours away from the ice and generally persist at this coverage for the remainder of the trajectory. After the crossover at five hours, these clouds maintain a cloud fraction on average 3% less than Towards clouds over the observation period. This difference between the non-MCAO and Towards clouds may be due to the fact that while non-MCAO clouds are, by definition, not exposed to the extremely powerful fluxes generated in MCAO events, they still move through less stable environments (higher MCAO, Figure 5.6 (h)) with higher windspeeds (Figure 5.6 (i)), promoting cloud formation by transporting energy and moisture from the surface to the boundary layer.

MCAO clouds sharply increase in cloud fraction within a few hours of leaving the ice edge, reaching coverage of about 95%. Between 7 and 40 hours TSI time, the MCAO cloud fraction decreases at approximately $-0.3\% \text{ hr}^{-1}$, with the cloud fraction falling below that of non-MCAO and Towards events at 14 and 22 hours, respectively. This decrease in cloud fraction is due to the stratiform layers initially produced in MCAO transitioning to the ‘roll-like’ cumulus clouds, commonly observed in outbreaks (Figure 5.1). The factors controlling the cloud fraction are discussed further in subsequent sections.

Liquid Water Path

The LWP of clouds moving towards the ice edge is generally steady, maintaining an average of around 80 g m^{-2} (Figure 5.6 (c)), which is in line with other liquid clouds in the region during spring to autumn (Shupe et al., 2006; Shupe, 2011). Non-MCAO clouds moving from the ice edge start with a lower LWP to these clouds, but overtake them at around 5 hours and remain on average 17 g m^{-2} higher for the rest of the trajectory, reaching a maximum of 100 g m^{-2} after 17 hours. For both of these sets of clouds, there is little variability in LWP along the trajectory.

In contrast, MCAO clouds show a rapid initial increase in LWP within 5-6 hours of leaving the ice, reaching a peak of around 127 g m^{-2} after 13 hours. The LWP then begins to change at a rate of $-1 \text{ g m}^{-2}\text{hr}^{-1}$, but remain higher than non-MCAO clouds for 40 hours. This initial increase and then decline with LWP along the trajectory has been found in other studies examining the evolution of MCAO cloud properties (Abel et al., 2017; Lloyd et al., 2018; Tornow et al., 2021). This LWP evolution has been attributed both to the instability and precipitation mechanisms in previous work (Tornow et al., 2021, 2022). The unstable conditions promote the entrainment of subsaturated air into the cloud, thereby reducing the LWP through evaporation (Chen et al., 2014; Michibata et al., 2016). Figure 5.6 (e) shows that precipitation is also key to LWP depletion, as the size of the droplets reach $15 \text{ }\mu\text{m}$, considered to be precipitation-sized (Rosenfeld and Gutman, 1994), just before the LWP declines. As with cloud fraction, a combination of effects related to the instability and precipitation mechanisms are explored in further sections.

Droplet Number Concentration

The N_d of clouds moving towards the ice edge increases over the trajectory, from about 200 cm^{-3} to 250 cm^{-3} (Figure 5.6 (d)). For non-MCAO clouds, the N_d values are similar to the Towards clouds close to the ice edge, but change at a rate -2 cm^{-3} hour and fall below the Towards clouds after 7 hours. For both of these trajectories, the values further from the ice edge are approximately the same or slightly higher than previous studies using MODIS have found (Zeng et al., 2014; McCoy et al., 2020), whereas the values close to the ice edge are higher. The larger N_d at the ice edge may be due to an increase in aerosol sources available in this biologically active zone (Leck and Persson, 1996), or the stronger winds (Figure 5.6 (h)) transporting aerosol to the cloud layer more efficiently. It is also possible that despite the extensive filtering of the satellite data, retrieval errors contribute to this increase near the ice edge. This extensive filtering has also likely biased the dataset used here relative to those used on other studies.

MCAO events start at lower N_d concentrations (200 cm^{-3}) and decline at a rate of $4 \text{ cm}^{-3} \text{ hr}^{-1}$ until about 25 hours into the trajectory. From there, the N_d concentration is relatively steady at about $90\text{-}100 \text{ cm}^{-3}$, which is on average 110 cm^{-3} lower than Towards clouds and 90 cm^{-3} lower than non-MCAO clouds. MCAO events may start at a lower N_d due to a potentially different origin of the air relative to non-MCAO events; the air may have travelled over ice for longer, and is therefore cooler and cleaner than the non-MCAO air. Previous work has found that some air masses can travel over the sea ice for extended periods before reaching the ocean (Silber and Shupe, 2022); however, a full investigation is beyond the scope of this study. Previous work has found a similarly steep decline in N_d concentration following an MCAO (Abel et al., 2017; Sanchez et al., 2022). Two main drivers of this N_d gradient in MCAO have been proposed; the dilution of aerosol concentration through

entrainment of air from the free troposphere and precipitation scavenging of aerosol from the below-cloud layer, which are considered further below.

TOA Albedo

For all sets of trajectories, the TOA albedo (Figure 5.6 (a)) generally follows the cloud fraction trend, as has been observed in previous studies (e.g Loeb et al., 2007; Bender et al., 2011). The TOA albedo for Towards trajectories is generally steady between 0.42 and 0.44, until very close to the ice edge where it increases relatively sharply to 0.46. This increase may be due to the increases in LWP and N_d close to the ice edge (Figure 5.6 (c) and (d)), despite decreases in cloud cover, or it may be due to a bias introduced by undetected sea ice. Although initially high, the MCAO TOA albedo gradually decreases from about 0.47 to 0.37 after about 35 hours, an albedo of about 0.06 below the non-MCAO and Towards clouds. Using CERES SYN1deg data, the average shortwave downwelling flux at 850 mbar (around the cloud top height; Figure 5.6 (g)) for the region of interest between March and October is calculated to be around 275 W m^{-2} . Therefore, the albedo difference means about 16 W m^{-2} more radiation reaching the surface relative to non-MCAO trajectories post-transition. However, this reduced cooling effect is only present during the summer, when the shortwave cooling effect of clouds is relevant.

5.3.2 Effects of instability

The intensity of MCAO events is expected to change as the high latitudes warm (Kolstad and Bracegirdle, 2008); therefore, it is important to know how the MCAO strength affects the evolution of cloud properties. Fletcher et al. (2016b) found that for mid-latitude outbreaks, clouds embedded in stronger MCAO events typically had

higher cloud fractions and optical thickness, which enhanced their shortwave effect. However, whether this relationship between MCAO strength and cloud properties persists over the temporal development of the cloud is uncertain.

To characterise the influence of MCAO strength on cloud development, the MCAO trajectories are divided based on whether their initial MCAO index (within the first 10 hours) fell into the upper or lower terciles of the M distribution (3.9 K and 1.5 K, respectively, similar to Fletcher et al. 2016b). As aerosols are also known to affect cloud development, the data are resampled such that the AOD distributions are equal for the strong and weak MCAO composites for each time step (following Gryspeerd et al., 2014). The resampling method involves dividing the AOD distribution at each time step into bins for strong and weak trajectories and randomly sampling the bin of the trajectory with more points in it until it matches the trajectory with fewer points (as illustrated in Figure 1 of Gryspeerd et al., 2014). This is repeated for each time step. The results are shown in Figure 5.7.

Cloud Fraction

Initially, clouds embedded in strong MCAO events have cloud fraction about 1-2% higher than those in weak events (Figure 5.7 (b)). The strong MCAO clouds maintain a high cloud fraction (94%) for several hours, but then decrease at a rate of $0.3\% \text{ hr}^{-1}$ after 7 hours. The weak MCAO cloud fraction begins to decline around the same time, at a similar rate ($0.2\% \text{ hr}^{-1}$). The gap in cloud coverage between the two trajectories closes at around 20 hours, after which the strong MCAO clouds are approximately 1% lower for the remainder of the observation period. Although small, the difference in the rate of decline results in weaker MCAO events having higher cloud coverage for longer; for instance, after 25 hours, the strong MCAO cloud fraction falls below 88% - a level only reached four hours later for weak MCAO

Investigating the development of clouds within marine cold air outbreaks

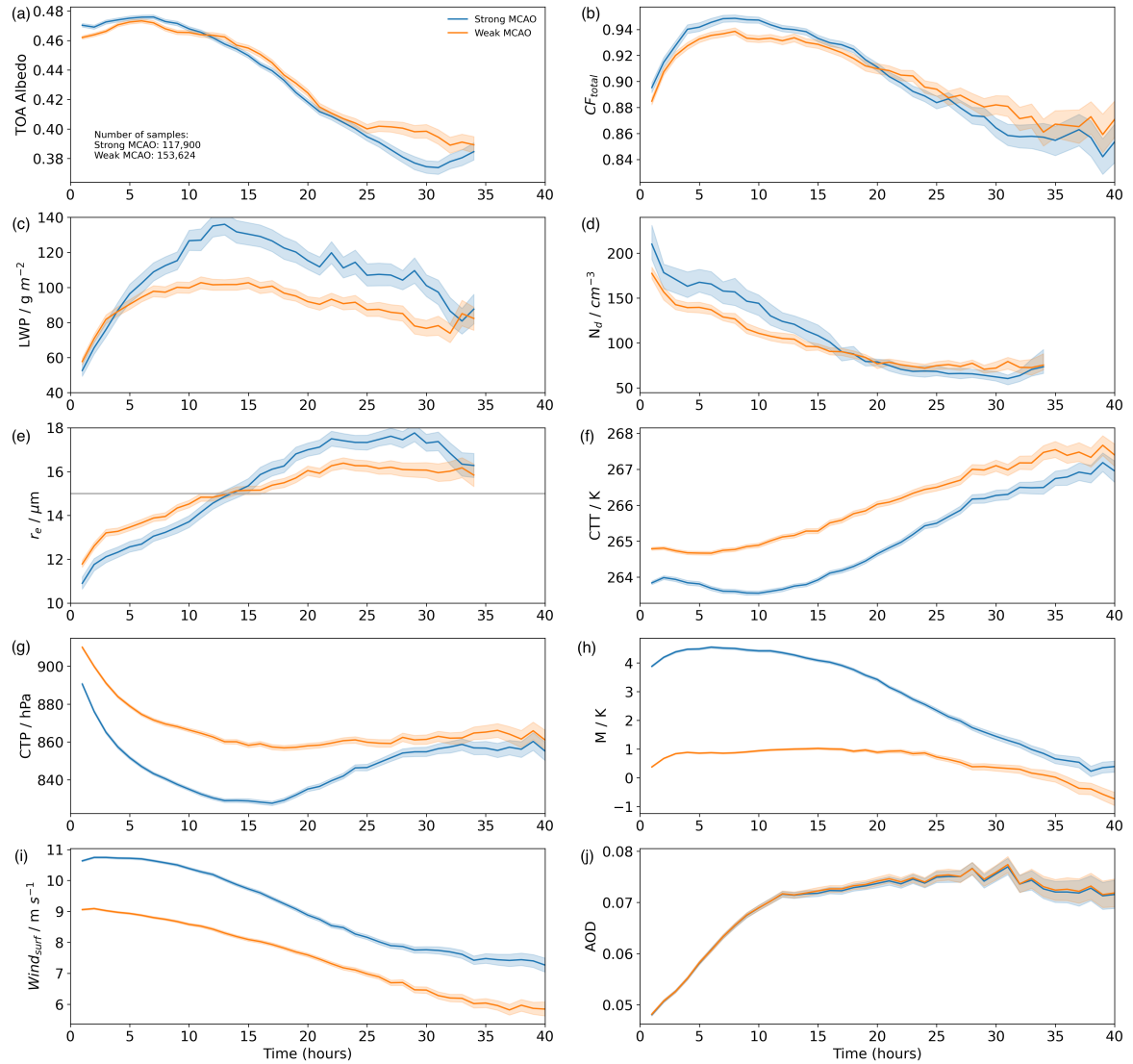


Figure 5.7: As Figure 5.6, but for strong ($M > 3.9$ K, blue) and weak ($0\ K < M < 1.5$ K, orange) outbreak events moving away from the ice edge.

events.

This faster decrease in cloud fraction for stronger outbreaks is mirrored by the N_d development (Figure 5.7 (d)). Clouds in strong MCAOs start with much higher N_d than those in lower intensity outbreaks, but fall steeply within the first 20 hours ($-7\ cm^{-3}\ hr^{-1}$), plateauing around $60-70\ cm^{-3}$. In contrast, N_d in lower MCAO events decreases more gradually over the first 24 hours ($-5\ cm^{-3}\ hr^{-1}$), steadying at about $10\ cm^{-3}$ higher than the strong MCAO case. These observations of cloud fraction

development concur with previous work, showing an increase in aerosol or N_d associated with an increase in cloud fraction (Gryspeerdt et al., 2016; Goren et al., 2019; Chen et al., 2022). As in Section 5.3.1, the cause for the difference in N_d between strong and weak events may depend on factors such as how long the air has spent over ice or the strength of the updrafts; while this is beyond the scope of this present study, it should be accounted for in future work investigating the effect of MCAO strength on cloud development.

The steeper decline in N_d (and therefore cloud fraction) in the stronger outbreaks is potentially due to the stronger entrainment of drier, cleaner air (Figure 5.8 (a)), leading to droplet evaporation and aerosol loss to the free troposphere (Tornow et al., 2022). The deeper boundary layer (implied by Figure 5.7 (g)) would also enhance decoupling from the surface, and therefore prevent the surface from acting as a source of moisture and aerosols, enhancing the decline in N_d , and hence cloud fraction. Another contributing factor to the more rapid N_d decline could be driven by collision-coalescence prior to precipitation; stronger outbreaks have higher LWP (Figure 5.7 (c)), which contributes strongly to the rain rate and accelerates collision-coalescence (Pawlowska and Brenguier, 2003), and therefore may reduce N_d . For both strong and weak events, precipitation is likely to be a factor in the very strong N_d decline early in the development. Although Figure 5.7 shows that the average droplet is below the commonly-used $15\mu\text{m}$ threshold for precipitation, Figure 5.8 (b) indicates that there are still a significant proportion of precipitation-sized droplets present in these clouds. It is also possible that undetected ice crystals may enhance precipitation and N_d loss through droplet riming (Tornow et al., 2021).

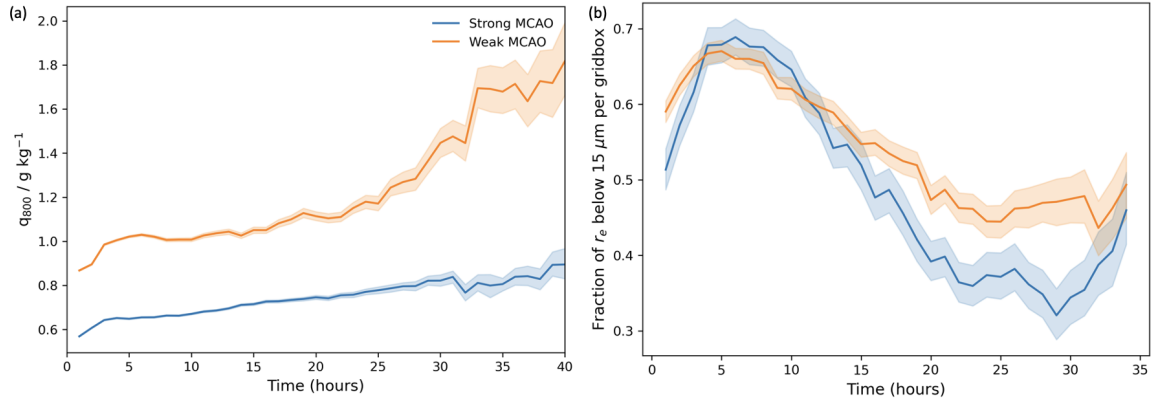


Figure 5.8: The specific humidity at 800 hPa (a) and fraction of pixels with r_e below $15 \mu\text{m}$ in a 25 km by 25 km gridbox (b) for strong ($M > 3.9 \text{ K}$, blue) and weak ($0 \text{ K} < M < 1.5 \text{ K}$, orange) outbreak events moving away from the ice edge.

Liquid Water Path

Instability has little relationship to LWP for the first few hours of cloud development (Figure 5.7 (c)). However, after 5 hours, LWP in strong MCAO events continue to increase, reaching a maximum of about 135 g m^{-2} at 13 hours. Following this peak, the strong MCAO LWP decreases at a rate of approximately $3 \text{ g m}^{-2} \text{ hr}^{-1}$. Conversely, the weak MCAO LWP remains around 100 g m^{-2} until 15 hours from ice, after which it decreases at a slower rate than the strong MCAO cases ($-1 \text{ g m}^{-2} \text{ hr}^{-1}$). Until around 33 hours, the weak MCAO cases maintain a LWP of about 20-30 g m^{-2} lower than the strong MCAO composite. Although entrainment drying would be enhanced in less stable conditions, the strong MCAO clouds have higher LWP to start with due to the heat and moisture fluxes associated with these events (Fletcher et al., 2016a), which also allow the clouds to grow deeper (implied by lower CTP in Figure 5.6 (g)).

For both sets of trajectories, the timing of the LWP decline is nearly coincident with the point at which the mean droplet reaches precipitation collision-coalescence threshold ($15 \mu\text{m}$, Figure 5.7 (e)), indicating the importance of precipitation to cloud break-up. Despite having an impact on the magnitude of the LWP increase, the MCAO strength does not appear to strongly modify the timing of the transi-

tion; the point at which LWP begins to decrease is only two to three hours earlier in the strong outbreaks. This may be due to the competing effects of LWP and N_d on precipitation (Goren et al., 2022); in strong MCAO events, LWP is high (which promotes precipitation), but higher N_d also suppresses precipitation. Conversely, in weaker events, lower LWP hinders precipitation while low N_d enhances precipitation formation. Therefore, the effects of LWP and N_d act as a buffer against each other in strong and weak MCAO events, leading to both trajectories reaching the 15 μm point at approximately the same time. It should be noted that the smaller droplet effective radius in clouds embedded in stronger outbreaks (Figure 5.7 (e)) is counter-intuitive, given these clouds are deeper than those in weaker outbreaks (Figure 5.7 (g)). This may be due to some unknown aerosol sources which are not well represented in the reanalysis data, meaning the attempts to constrain the effects of aerosols are not fully effective.

TOA Albedo

Despite the differences in the evolution of the macro- and micro-physical properties, there is a negligible difference (0.01-0.02) between the scene albedo over the course of the strong and weak MCAO trajectories. This may be due to the competing effects on the albedo; although the cloud fraction of the strong events does fall slightly below the weaker events over time, the enhanced LWP compensates for this decline, leading to roughly similar reflectivities. This suggests that should the MCAO strength change with the changing climate (e.g., Kolstad and Bracegirdle, 2008), the impact on the shortwave energy budget due to changes in cloud properties may be minimal.

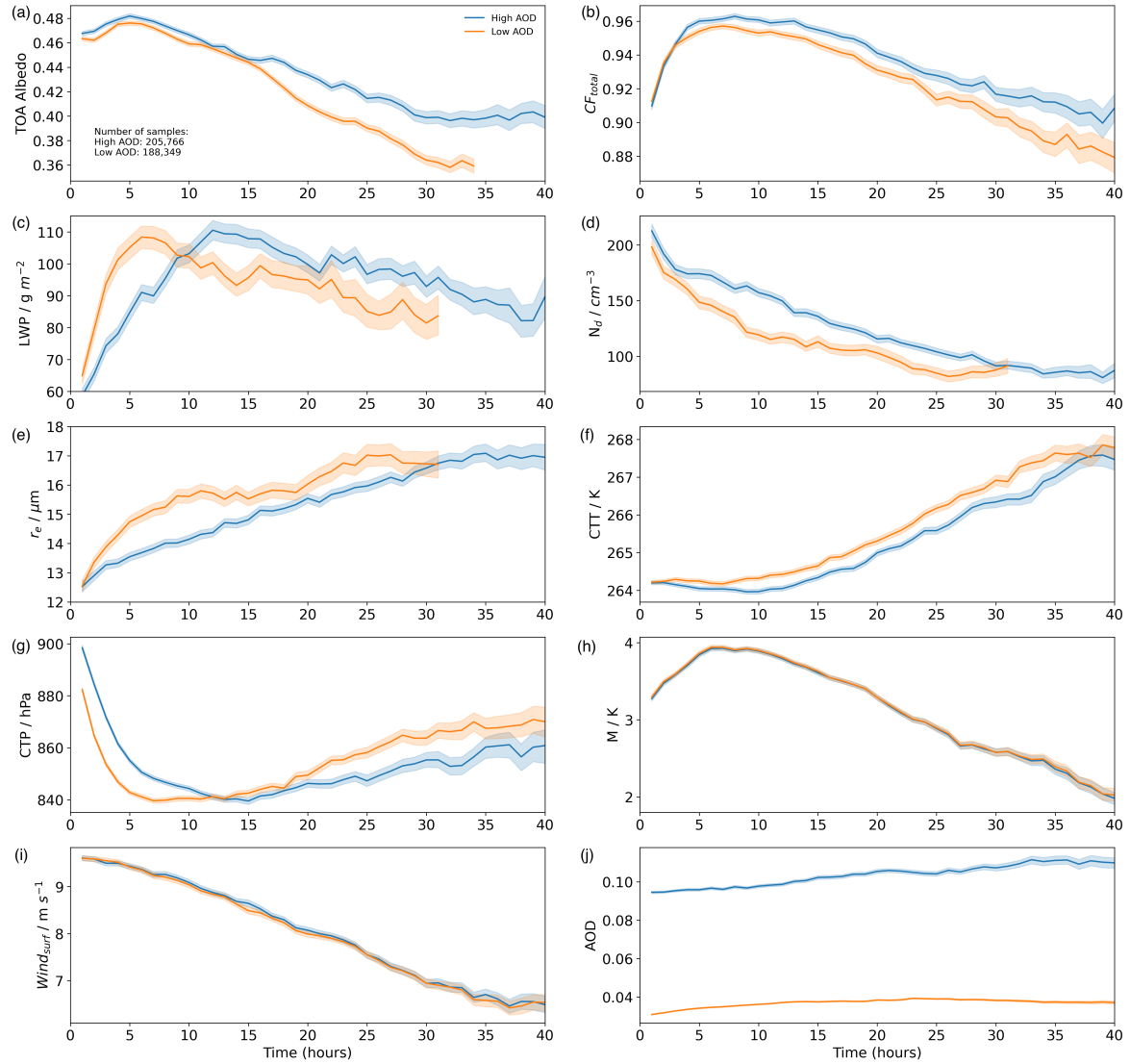


Figure 5.9: As Figure 5.6, but for trajectories in high and low aerosol conditions.

5.3.3 Effects of aerosol on cloud development

Aerosols can affect the transition from stratocumulus decks to broken cloud fields influencing the timing of precipitation (e.g Christensen et al., 2020). In MCAOs, higher aerosol loads can delay the formation of the cumuliform regime (Tornow et al., 2021, 2022). However, once precipitation occurs, the aerosol scavenging accelerates the cloud field's break-up through enhanced water loss from the stratocumulus layer (Abel et al., 2017; Lloyd et al., 2018). The timescales over which aerosols modulate MCAO cloud development have not previously been studied with observational data.

To characterise these timescales, the MCAO trajectories are divided into above and below median (0.06) aerosol conditions based on the AOD early in their trajectory (within the first 10 hours). As previously discussed, AOD from reanalysis is imperfect, especially in the Arctic, with aerosol sources potentially missing from the data product. Furthermore, the vertical profile of the aerosol load is important to clouds in MCAO; Tornow et al. (2022) showed that entrainment of free tropospheric air may reduce the aerosol burden in MCAO. As AOD is a column-integrated value, it does not describe the aerosol concentration throughout the cloud. Additionally, the AOD does not differentiate between INP and CCN, which is important considering the differing roles they play in cloud lifetime. However, INP concentrations are typically less abundant than CCN by several orders of magnitude (e.g., Bigg and Leck, 2001). Here, the AOD is used qualitatively to indicate whether the trajectories are moving through more or less aerosol-laden conditions. CAMS characterises these aerosols as either sulfate or sea salt (not shown).

The influence of boundary layer instability on cloud development is constrained using the resampling method discussed in Section 4.2 on M for each time step. Additionally, as the wind speed is also known to affect cloud properties (such as cloud fraction; Engström and Ekman 2010), it is also resampled to be the same for the high and low AOD trajectories. The M and wind speed distributions are resampled sequentially. In this work, M is resampled before the wind speed, but as the two are highly correlated the order of which variable is resampled first has little effect on the results (not shown).

Cloud Fraction

Figure 5.9 (b) shows that there is initially a small difference in cloud fraction between the high and low AOD cases. However, after about three hours, the effects of aerosol become apparent and the trajectories diverge. The high AOD cases reach 96% coverage, and maintain this level until about 10 hours from ice, after which it decreases at a rate of about $0.2\% \text{ hr}^{-1}$. The low AOD cases remain about 1-2% lower, with the difference growing over time. As before, the cloud fraction evolution is mirrored by the N_d development in both high and low AOD conditions. Initially, the N_d for both trajectories is similar, but the decrease in N_d in cleaner conditions is more rapid than in cases with more aerosol ($-6 \text{ cm}^{-3} \text{ hour}^{-1}$ versus $-4 \text{ cm}^{-3} \text{ hour}^{-1}$).

As M and wind speed are accounted for by the resampling, both the dilution of aerosol to the free troposphere and ability of the surface to act as a source is similar in both high and low AOD cases. Therefore, the difference between the two sets of outbreaks is likely due to enhanced precipitation earlier in the low aerosol trajectories. Figure 5.9 (e) shows that the cloud droplets grow in size more quickly in cleaner conditions, and reach precipitation-sized after about 6-7 hours. Higher aerosol loads mean cloud droplets stay smaller for longer, keeping the N_d higher and maintaining a higher cloud coverage for a longer time.

Liquid Water Path

In contrast to the effects of outbreak strength, which strongly modulated the peak LWP but not the timing, changes in aerosol conditions influence the point of LWP decline but only moderately affect peak LWP magnitude (Figure 5.9 (c)). Clouds in low AOD trajectories have a much earlier peak in LWP (about 108 g m^{-2} at 5-7 hours) than clouds in high AOD trajectories (112 g m^{-2} at 12 hours). The LWP decreases

in both sets of trajectories are closely linked to precipitation dynamics; the point at which r_e reaches $15 \mu\text{m}$ is within 1-2 hours of the start of the LWP decline in both cases. As with the cloud fraction cases, more aerosols lead to precipitation suppression, which allows the LWP to build up more gradually. However, due to the build up of cloud water, precipitation eventually still occurs, triggering the transition. The near doubling of the time to the LWP peak highlights the potentially significant effect that aerosols, through influencing precipitation dynamics, can have on the cloud development.

TOA Albedo

As would be expected from the cloud fraction and LWP development, the TOA albedo declines more slowly in high AOD trajectories. Although the difference in albedo is initially small, the composites diverge over time, eventually growing to 0.04 after about 30 hours. This approximately corresponds to an additional 11 W m^{-2} (using the regional average shortwave downwelling radiation at 850 mbar) of cooling in summer due to the prolonged high cloud cover and delayed LWP peak. This cooling is not insignificant; Huang et al. (2017) found that anomalies in the shortwave cooling of a similar magnitude caused by changes in cloud properties during late spring and early summer influenced the extent of sea ice melt.

5.3.4 Stability-dependent aerosol effect

Section 5.3.3 considered the effects of an aerosol perturbation while constraining the instability; however, previous work has shown that the cloud response to aerosols can change depending on the stability environment (Chapter 4). Therefore, the response of cloud properties to aerosols may depend on the strength of the MCAO.

Figure 5.10 shows the effects of dividing the MCAO trajectories into four regimes, strong/weak MCAO and high/low aerosol, based on the upper and lower M terciles and median AOD, as in previous sections. To highlight the overall differences in responses between clouds in strong and weak events, M is constrained to be identical along high/low aerosol trajectories for each case.

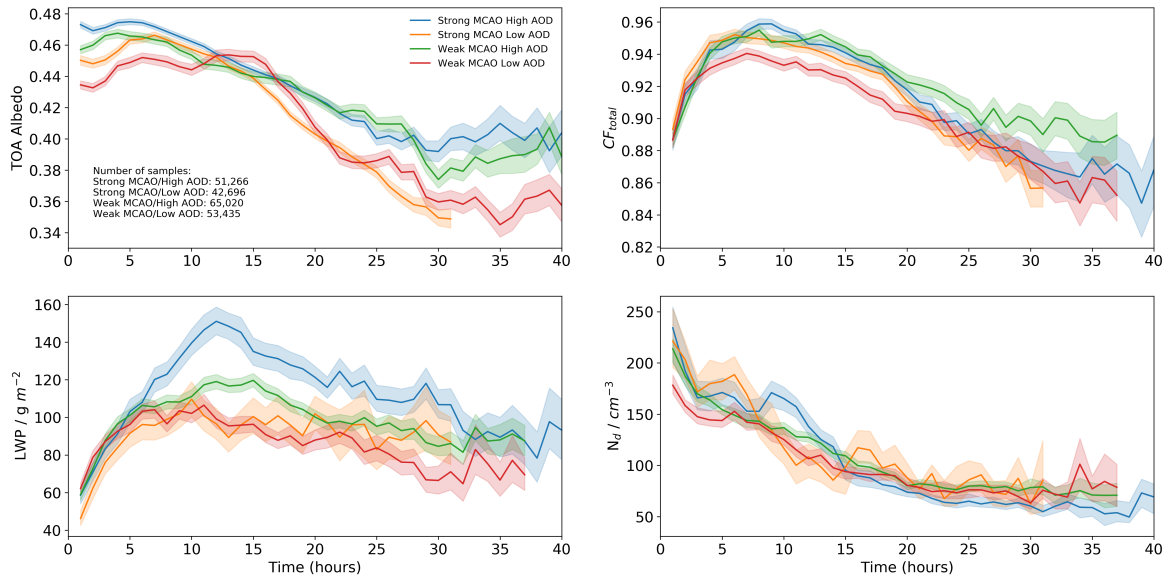


Figure 5.10: As Figure 5.6, but for trajectories in strong/weak MCAO and high/low aerosol conditions.

In general, the effect of aerosols on clouds in strong and weak events is similar to that seen in Section 5.3.3; clouds in higher aerosol conditions typically have higher cloud fractions (Figure 5.10 (b)) and delayed peak in LWP (Figure 5.10 (c)), leading to a higher albedo (Figure 5.10 (a)). In strong MCAO events, the difference in albedo between the high and low aerosol conditions appears to be driven by the large difference in LWP, with relatively little difference in cloud fraction between the two cases. However, both the cloud fraction and the LWP contribute to the higher albedo in the weak MCAO/high AOD cases. Previous work has shown that in relatively clean conditions, as the aerosol load increases, less stable conditions typically have a higher LWP than stable conditions (Chapter 4), potentially explaining the difference

in LWP response between strong and weak events in Figure 5.10 (c). However, it is unclear why the aerosol load does not strongly affect the cloud coverage in strong MCAO events; the large surface fluxes may be the dominant term in promoting cloud formation, leaving little sensitivity to aerosol.

5.4 Discussion

This work presents a novel way of investigating the temporal development of cloud properties, using cold air outbreaks as case studies. This is done by creating composite trajectories and co-locating these with satellite and reanalysis data. Through comparison with clouds in non-MCAO events, the extent to which the extreme turbulent fluxes affect the cloud development and formation are revealed. Many of the results here confirm what has been observed in model results or field campaigns; deep, high-coverage clouds form quickly as the air moves from the ice edge. Eventually, through precipitation, these clouds transition into thinner, low coverage cloud fields. The cloud microphysical properties also evolve throughout the outbreak; N_d declines and LWP initially builds up before decreasing. The unique aspect of this study is the insight into the timescales for these processes occurring, and the ability to use reanalysis data to probe the mechanisms controlling these cloud properties.

When considering the effects of MCAO strength, these composite trajectories reveal the importance of considering how far the clouds are along the outbreak. Fletcher et al. (2016b) found that clouds in stronger outbreaks had higher cloud fraction and optical thickness, and therefore a greater shortwave cooling effect. However, weak and strong outbreak cases were selected based on the value of M for a given gridbox. Figure 5.7 (g) shows that M decreases over the course of a trajectory, particularly for strong events, so that selection by gridbox is actually choosing clouds at different

points in their development. Although initially lower, weak outbreaks actually maintain a higher coverage for longer. However, due to the enhanced LWP in stronger outbreaks, this does not translate to a strong change in TOA albedo or shortwave cooling effect of the cloud.

Due to changes in the Arctic climate, the strength and frequency of MCAO are expected to change in the future. Several modelling studies have projected an overall decrease in the M, except in areas of sea ice retreat, where M will increase due to the availability of the ocean surface (Kolstad and Bracegirdle, 2008; Landgren et al., 2019). Figure 5.7 suggests that a shift in the MCAO strength would not strongly shift the shortwave cooling effect of the clouds, due to the competing factors controlling the TOA albedo. However, should the MCAO weaken so much that they constitute ‘non-MCAO’ events moving off the ice edge, there would be a shift to higher-albedo, more cooling clouds over their trajectories (Figure 5.6 (a)). The data presented here only considers MCAO events during sunlit months, where the shortwave cooling effect of low-level clouds over the ocean surface is expected to be dominant over the longwave warming. However, MCAOs are most common during winter (Fletcher et al., 2016a), when a lack of sunlight precludes the analysis of some cloud properties. Although Fletcher et al. (2016b) found that MCAO strength, rather than season, was the strongest determinant of cloud properties, the generalisability of the results presented here is uncertain. However, if these results hold, it would be the longwave warming effect of the clouds which is most relevant. As the clouds in this study typically have an emissivity near unity (for $LWP > 30 \text{ g m}^{-2}$; Shupe and Intrieri 2004), the cloud longwave effect is predominantly controlled by cloud cover. If the MCAO-CF relationship holds in a future Arctic, this suggests that a weakening of the MCAO would generate longer-lasting, more warming clouds.

As more aerosol sources such as industry or shipping move further into the Arctic,

from Figure 5.9 it appears that should these aerosols interact with clouds in MCAO, they would lead to longer-lived stratocumulus fields, and these higher-coverage, thicker clouds would have either a stronger warming or cooling effect, depending on the season. These results highlight that knowledge of the aerosol environment close to the ice edge is essential to model the development of these clouds correctly. Despite recent efforts, such as the measurement of aerosols capable of ice nucleation during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAIC) expedition (Creamean et al., 2022; Shupe et al., 2022), the aerosol sources and sinks close to the ice edge are not well characterised, leaving models poorly constrained. Given the effects that aerosols have on the timing of the cloud transition, this uncertainty around aerosol sources may make it difficult to determine the radiative impact of these clouds. The importance of understanding aerosol conditions in the Arctic are discussed further in Chapter 6.

While this work considers liquid-dominant clouds, mixed-phase clouds often form in MCAO. As seen in Chapter 3, the misclassification of ice may introduce bias in the microphysical property measurements. It may be expected that detecting an ice crystal as a liquid drop may produce a higher measurement for the droplet effective radius due to the different scattering and absorption properties of ice crystals (Platnick et al., 2003). This would lead to an underestimation in N_d and overestimation of LWP. Figure 5.7 (e) shows that stronger events - typically associated with more ice (Fletcher et al., 2016b) - have a lower r_e than lower MCAO clouds, suggesting that this is not the dominant effect. However, it is possible that ice undetected by MODIS has a non-negligible influence on these results.

5.5 Conclusions

Clouds in marine cold air outbreaks undergo a characteristic evolution, transforming from high-coverage, stratiform clouds to a broken, low-coverage cumuliform cloud field. Despite previous flight and measurement campaigns, models struggle to capture this development due to the complex set of factors controlling cloud properties. This work uses a set of composite trajectories to develop a time-resolved picture of how cloud properties change as air moves away from the ice edge and the effect of different environmental conditions on the cloud evolution.

MCAO cloud properties are distinct from clouds following other, non-MCAO trajectories (Figure 5.6 (b) - (e)). The cloud albedo is largely controlled by cloud fraction, so as the clouds transition from high- to low-coverage cloud fields during an outbreak, their shortwave cooling effect weakens. The cloud fraction in turn is strongly correlated with the N_d , a relationship seen in previous studies and which holds across the range of factors considered here. Similarly, along each set of composite trajectories, the timing of LWP peak is nearly coincident with the point at which the mean r_e reaches $15\text{ }\mu\text{m}$, indicating the importance of precipitation to cloud transition.

The initial MCAO strength has a lasting impact on the cloud property development, particularly visible in the LWP (Figure 5.7 (c)). Previous work investigating the impact of MCAO strength on clouds has considered a gridbox-by-gridbox approach, rather than how clouds develop as the outbreak progresses. These results show that for some cloud properties (such as the cloud fraction, Figure 5.7 (b)), clouds in strong MCAO events typically start with a higher values, as had been seen in previous studies, but this does not hold at later stages of development. However, despite differences in cloud property development, the opposing influences of the cloud fraction and LWP development mean that the TOA albedo for clouds in each

set of composite trajectories is very similar (Figure 5.7 (a)). Furthermore, the timing of the transition is not strongly changed by the MCAO strength.

In contrast, a higher initial aerosol load delayed the transition by 5-7 hours and slowed the decline in cloud fraction, enhancing the TOA shortwave cooling by approximately 11 W m^{-2} relative to low aerosol conditions (Figure 5.9). This suggests that in a future, more polluted Arctic, these new aerosols sources may increase the cloud cooling effect through delaying the stratocumulus-to-cumulus transition. This potentially strong impact on cloud development and radiative properties highlights the need to constrain sources of Arctic aerosols, particularly close to the ice edge.

This work solely focuses on liquid-dominated clouds embedded in MCAO, which limits the study to only a third of all available data. However, mixed-phase clouds are a common feature of these events, and the formation of ice can be important to the development of other cloud properties. For example, ice crystals may rime the liquid droplets, enhancing precipitation and contributing to the steep N_d gradient in Figure 5.7 (d). In general, mixed-phase clouds are challenging to model due to the complexity of physical processes associated with their formation and maintenance and a lack of observations of fundamental mechanisms, which lead to uncertain parameterisations (Morrison et al., 2012). Chapter 6 will extend the TSI method to investigate the development of ice-containing clouds in MCAO. There will be a focus on examining the impact of air mass history on mixed-phase cloud formation, with an emphasis on INP availability.

Chapter 6

Investigating the links between air mass history and the development of mixed-phase clouds

Key Points

- Clouds formed in MCAOs are characterized by a rapid shift from liquid to ice-containing states, contrasting with other clouds in the region.
- Mixed-phase clouds in MCAOs initially form predominantly at temperatures below -20°C but later become dominant at temperatures as high as -13°C , hinting at the role of biological INPs.
- The evolution of mixed-phase clouds during MCAOs is significantly influenced by the air mass history, particularly the duration spent over ice- and snow-covered surfaces, which are typically deficient in INPs.
- Seasonal variations in cloud development are observed both within MCAOs and in non-MCAO events, linked to differences in air mass history and INP availability.

- Mixed-phase clouds are observed to form as high as -5°C later in MCAO events, with some evidence suggesting secondary ice production as the cause.
- This study highlights the need for more detailed observations and understanding of aerosol sources in the Arctic, especially near the ice edge, and the mechanisms of aerosol transport to the cloud layer.

The contents of this chapter are based on Murray-Watson, R. J. and Gryspeerdt, E.: Air mass history linked to the development of Arctic mixed-phase clouds, EGUsphere [preprint], <https://doi.org/10.5194/egusphere-2024-129>, 2024. The analysis and text are my own work, with comments from my co-author.

6.1 Introduction

As discussed in Chapter 5, understanding the factors influencing precipitation formation is key to understanding cloud life cycles. Ice formation is hypothesized to enhance precipitation, and may accelerate the transition from stratocumulus to cumulus clouds in MCAOs (Tornow et al., 2021). However, the factors which affect the formation and evolution of ice in these clouds are highly uncertain, in part due to a lack of observations in the region. This chapter uses the TSI method to characterise the evolution of mixed-phase clouds formed in MCAO, and expands on this method to consider the influence of air mass history on their formation.

Some previous modelling work has highlighted the role of INP in MCAO; for example, Vergara-Temprado et al. (2018) found prolonged stratocumulus coverage under low INP conditions. High INP concentrations resulted in broken cumulus clouds, leading to significant reductions in the shortwave cooling. Flight campaign mea-

measurements of clouds in MCAOs indicate that there is typically little ice in the stratocumulus regime and substantially more in the cumulus clouds (Abel et al., 2017; Lloyd et al., 2018). However, this shift to a more glaciated cloud regime cannot be entirely explained by the presence of INP; Abel et al. (2017) observed a rapid enhancement of ice crystal number concentration that was not matched by the ambient INP availability. Therefore, secondary ice production, in which an ice crystal is formed from an existing ice crystal, might be necessary to explain the development of ice-containing clouds observed in these outbreaks.

One of the most well-known secondary ice production processes is the Hallett-Mossop mechanism. This is the process by which ice splinters are produced following the riming of a supercooled droplet onto an existing ice particle (Hallett and Mossop, 1974). These ice splinters themselves can rime onto droplets and produce more ice, therefore reducing the dependence on INP for ice formation. The process is most efficient between -3°C and -8°C and for droplets with a diameter greater than $24\text{ }\mu\text{m}$ or less than $13\text{ }\mu\text{m}$ (Mossop, 1978, 1985; Heymsfield and Mossop, 1984). The enhanced ice crystal concentration observed in Abel et al. (2017) occurred in lower-level cumuliform clouds which had cloud base temperatures within the Hallett-Mossop temperature range, suggesting that this mechanism was active. Through modelling, they found that this enhanced precipitation from this secondary ice production was essential for the stratocumulus-to-cumulus transition. Surface-based radar observations from the Cold-Air Outbreaks in the Marine Boundary Layer Experiment (COMBLE) field campaign (Geerts et al., 2022) also found evidence of secondary ice production in MCAO clouds in the Hallett-Mossop temperature range, with ice multiplication factors on the order of 100 (Mages et al., 2023). Although there is still uncertainty about the physical mechanism of the Hallett-Mossop process, it is one of the only secondary ice production mechanisms parameterised in weather and climate models.

Riming, or the freezing of supercooled droplets onto ice crystals, is also thought to influence MCAO transitions. Abel et al. (2017) found rimed graupel particles in both the pre-and post-transition clouds. Tornow et al. (2021) suggested a mechanism called ‘preconditioning by riming’, during which the formation of ice precipitation primes the MCAO clouds for transitioning. Through reducing the cloud water, scavenging aerosols and cooling the sub-cloud layer, increasing ice concentrations accelerated the cloud break-up.

Even given the importance of secondary ice production, it is evident that sources of INP in the region have a strong control on MCAO cloud development. However, there is great uncertainty about INPs globally due to the diversity of sources, variety of physicochemical properties, low concentrations and seasonal cycle (Murray et al., 2012, 2021). This is particularly true in the Arctic, where INP concentrations are much lower than the global average (Li et al., 2023), and sparse measurements contribute to a greater lack of knowledge (Schmale et al., 2021). However, a number of field campaigns or in situ measurements have identified the Arctic ocean as a source of biological INP (Bigg, 1996; Bigg and Leck, 2001; Irish et al., 2017, 2019; Creamean et al., 2019; Hartmann et al., 2021; Porter et al., 2022; Li et al., 2023). This organic matter is transported from the sea surface to the atmosphere via a bubble-bursting mechanism (Wilson et al., 2015). Wex et al. (2019) identified a seasonal cycle to INP availability around the Arctic, with higher concentrations in the summer months, likely due to enhanced biological activity spurred by increased availability of sunlight. Biological INP are distinctive as they can cause ice formation at higher temperatures than other INP such as mineral dust, and are typically the dominant INP active above -15 °C.

Several studies have explored the influence of long-distance transport and the length

of time an air parcel remains over various surface types on INP present in those air masses. Porter et al. (2022) found that in the summertime high Arctic, the presence of high-temperature INP was associated with air parcels that had spent time over land or ocean. In contrast, time over the sea ice was associated with low aerosol concentrations (Mauritsen et al., 2011). Similarly, Irish et al. (2019) found a positive correlation between INP concentration and time over snow-free land surface and a negative correlation with time spent over pack ice. However, they found a weakly negative relationship between INP concentrations and time spent over the open ocean. Li et al. (2023) also found low INP populations in air masses which travelled over the sea ice, and that long-range dust from Arctic sources such as Greenland was associated with particularly high INP concentrations. Bigg (1996) linked the time since the air parcel had been over the open ocean with declining INP concentrations, suggesting that the marine sources dominated INP concentrations. In contrast to many of these studies, Hartmann et al. (2021) found no connection between INP concentrations and air mass history, and determined that the observed marine biological INP were produced locally.

As previously discussed, MCAOs have proven a challenge to model due to the difficulties in capturing the complex system dynamics. Improving the cloud microphysics scheme, in particular the ice formation processes at warmer temperatures, can lead to a better representation of MCAO clouds, but biases still exist (Field et al., 2014). Poor representation of ice processes in models causes the rapid depletion of liquid, leading to an earlier breakdown of stratocumulus clouds than observed and lower amounts of supercooled water (Abel et al., 2017; Van Weverberg et al., 2023). In the Southern Ocean, the inability to represent supercooled water in MCAO clouds has been identified as a central factor contributing to biases in shortwave radiation (Bodas-Salcedo et al., 2014, 2016). In general, mixed-phase clouds are challenging to model due to the complexity of physical processes associated with their formation

and maintenance and a lack of observations of fundamental mechanisms, which lead to uncertain parameterisations (Morrison et al., 2012). Similarly to the MCAO-specific cases, Cesana et al. (2022) found that GCMs which have more complex microphysics schemes typically produce better simulations of the liquid-to-ice ratio in mixed-phase clouds in the Southern Ocean than simpler schemes. As mixed-phase clouds are central to the Arctic energy budget (Shupe and Intrieri, 2004) and may prove important components of the cloud-radiative feedbacks (Tan and Storelvmo, 2019), targeting the uncertainty surrounding them is key for improving future projections of the Arctic climate.

It is evident that an advanced understanding of the formation of ice in cold-air outbreak clouds is required to improve the modelling deficiencies. There have been several observational campaigns targeting cold-air outbreak clouds, such as the recent COMBLE campaign in the north European Arctic (Geerts et al., 2022) and HALO-(AC)³ flight campaign. However, many of the observational studies are limited in space or time, and therefore provide a relatively limited picture of the cloud development. Satellites can overcome some of these challenges, and while they do not provide as high-resolution data as the in situ studies, they offer the opportunity to capture the spatial and temporal dynamics of cold-air outbreak clouds on a much larger scale. Previous studies typically have relied on snapshot satellite imagery to investigate the prevalence of ice-containing clouds in MCAOs (e.g., Fletcher et al., 2016b), despite these properties depending on the stage of the outbreak’s progression, as ice is more likely to be found in post-transition cumuliform clouds (Abel et al., 2017).

This work targets the uncertainty surrounding the development of mixed-phase clouds in MCAOs. In particular, it addresses two aspects of mixed-phase development; the temporal scales over which the mixed-phase clouds form and the depen-

dence on temperature, and the role that air mass history may play in the types of INP available for ice formation. The TSI trajectories described in Chapter 5 are used with active remote sensing data to create a composite picture of temporal development of mixed-phase during these outbreaks. As in Chapter 5, these are compared to ‘non-MCAO’ clouds to discern the influences on general mixed-phase clouds in the region. To investigate the relative importance of local aerosols versus distant INP sources, the influence of origin of the air mass and the types of surfaces it has passed over before reaching the pack ice on mixed-phase cloud formation are examined.

6.2 Methods

A detailed description of the TSI method is given in Section 5.2.2. This chapter also considers an extension to this method by examining the time a given air parcel has spent over the sea ice and snow-covered surfaces before it leaves again and moves over the open ocean. Previous work suggests that the duration spent over the sea ice can change the air mass properties and the types of clouds formed (Silber and Shupe, 2022). In the high Arctic, the boundary layer above the pack ice can be extremely clean, with measurements of CCN concentration recorded below 1 cm^{-3} during the Arctic Summer Cloud Ocean Study (ASCOS) campaign (Mauritsen et al., 2011). The method for ‘time over sea ice/land’ is similar to the TSI method, but instead considers all pixels above 50°N in order to better capture the air mass history, including air moving into the Arctic region. The surface type data is obtained from the Interactive Multisensor Snow and Ice Mapping System (IMS) Daily Northern Hemisphere Snow and Ice Analysis at 1km Resolution, Version 1 product (U.S. National Ice Center, 2008), and is regridded to the same polar stereographic, 25 km by 25 km grid as the sea ice data. The ‘Land Without Snow’ and ‘Snow Covered Land’ were used to define ‘land’ pixels. If a pixel contains a mix of surface types, it

is categorized under the dominant type when this type occupies more than 75% of the pixel area. Otherwise, the pixel is characterised as ‘uncertain’.

The reanalysis products, namely the wind data and the fields used to calculate the MCAO index (M), were processed as in previous chapters.

6.2.1 Cloud Properties

Cloud phase data were obtained from DARDAR v2 (Delanoë and Hogan 2010; Ceccaldi et al. 2013), and were processed as described in Section 2.3. Cases where the cloud top height is below 720 m are excluded due to surface clutter affecting the radar retrievals, as in other studies (e.g., Kay and Gettelman, 2009). Clouds with cloud top heights recorded above 5000 m are excluded to limit the inclusion of high-level clouds not associated with MCAOs. Although this may exclude some MCAO clouds, other work has found cloud tops are typically much lower than this (Fletcher et al., 2016b; Lackner et al., 2023; Mateling et al., 2023).

Cloud effective radius (r_e) measurements were obtained from MODIS, and processed as described in Chapter 4.

6.3 Results

6.3.1 Phase evolution as a function of time since the ice edge

Clouds created in MCAOs have a distinctive phase evolution relative to other clouds in the region (Figure 6.1), most notably from the ice- and mixed-phases. MCAO clouds are characterised by the rapid decline of the liquid phase down to about 20%

after 6 hours, with a corresponding increase in mixed-phase clouds. While the liquid fraction then remains relatively steady, the mixed-phase proceeds to glaciata and convert to ice-only clouds. In contrast, for both non-MCAO and Towards clouds, the liquid phase remains dominant. The phase fractions remain relatively steady as non-MCAO clouds move off the ice edge, with nearly twice as much liquid cloud as mixed-phase and very few (about 3%) ice clouds. Although the composition of Towards clouds also shifts towards ice-containing phases as they move further north, the shift is very gradual, and liquid clouds are still dominant. This shift to mixed-phase is likely in part due to decreases in cloud temperature as the clouds move towards the ice. Additionally, as these clouds move north from lower latitudes, the air masses may potentially have more INP than the air which has spent greater lengths of time over the sea ice (discussed more in Section 6.3.3), and therefore more readily form mixed-phase clouds. Broadly, the dominance of liquid-containing phases in the Towards clouds agrees with previous climatologies of low-level Arctic clouds (e.g., Shupe et al., 2006; Cesana et al., 2012; Yan et al., 2020), but does show more liquid-only clouds than some other studies (e.g., Shupe, 2011), although this may be due to different study regions. Using DARDAR, Fletcher et al. (2016b) also determined that clouds within outbreaks had higher ice fractions compared to other regional clouds.

This shift in MCAO clouds towards more glaciated state is in agreement with previous observations; Abel et al. (2017) found much larger quantities of ice in the post-transition cumulus clouds relative to the upstream stratocumulus deck. The following sections will explore the potential roles of the air mass history and the cloud top temperature in influencing the observed disparities in ice-containing phases between MCAO clouds and non-MCAO trajectories.

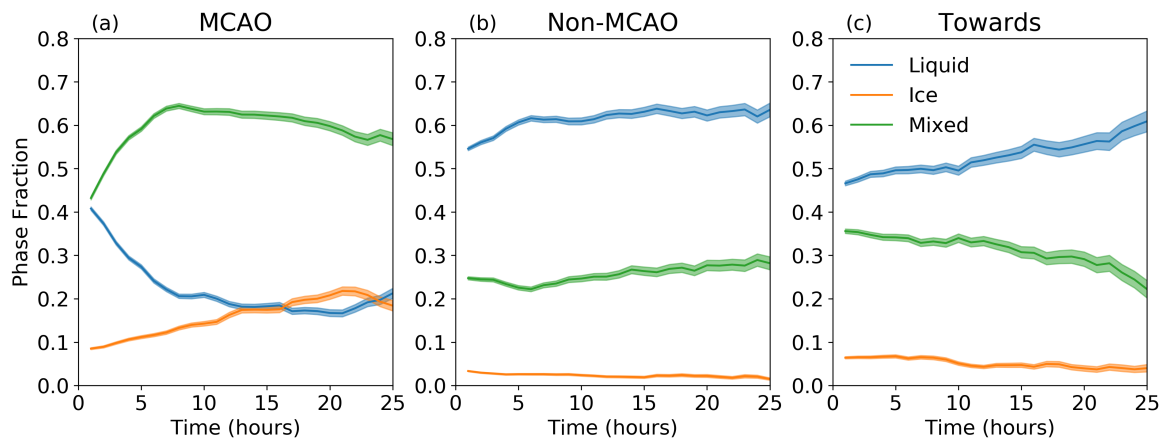


Figure 6.1: Evolution of cloud phase as a function of time for (a) MCAO, (b) non-MCAO and (c) Towards trajectories.

6.3.2 Mixed-phase development as a function of temperature and time since ice

Cloud top temperature is an important factor in the formation of ice in clouds (e.g., Korolev et al., 2003; Shupe et al., 2006; Westbrook and Illingworth, 2011; de Boer et al., 2011), typically with increasing amounts of ice at lower temperatures. However, supercooled liquid clouds have also been observed to exist at temperatures below -15°C (e.g., D'Alessandro et al., 2021; Danker et al., 2022; Carlsen and David, 2022). Figure 6.2 shows the evolution of the mixed-phase fraction for MCAO and non-MCAO clouds as a function of the time since ice and cloud-top temperature. Mixed-phase clouds are initially only dominant at very low temperatures (-23°C) in MCAO. As the clouds progress from the ice edge, the temperature at which the majority of clouds are mixed-phase increases, until a plateau at approximately -13°C after 7 hours. The low mixed-phase fraction at very low temperatures further into the trajectories (bottom-right of Figure 6.2 (a)) is due to the shift to ice-only clouds (Figure 6.3).

This is in contrast to the non-MCAO clouds; from the point of leaving the ice edge onwards, mixed-phase clouds are prevalent below -13°C and supercooled liquid domi-

nant at warmer temperatures. Figure 6.2 (c) underlines the differences in this initial development by emphasising the initial relative lack of mixed-phase clouds early in MCAO (the gradient highlighted as Region A). This gradient in the temperature at which mixed-phase clouds become the majority has previously been observed by Carlsen and David (2022), who found that the temperature at which a cloud regime switched from being liquid-dominant to mixed-phase dominant (which they called T^*) increased as a function of distance from the ice edge, until a plateau is reached at about -15°C . They also observed a seasonal dependence; during the summer, there was a slightly weaker gradient in T^* from the ice edge (from about -17°C to -15°C) than in the winter (from about -22°C to about -15°C). They attributed this difference due to INP abundance, but it may also be due to the seasonal differences in MCAO events; there are relatively few MCAO in summer (Figure 6.4), so they may have been observing events more similar to Figure 6.2 (b), i.e events which are moving off of the ice edge, but are not necessarily MCAO. Conversely, in winter, their observations match Figure 6.2 (a) more closely. The seasonal dependence of these MCAO and non-MCAO cloud evolution is discussed further in Section 6.3.3.

Figure 6.2 (a) also reveals another interesting feature of the MCAO clouds; after about 7 hours, a significant proportion of the clouds are mixed-phase at higher temperatures (between -13°C and -3°C). On average, the mixed-phase proportion at these temperatures in MCAOs is 15-20% greater than in non-MCAO clouds (Figure 6.2 (c), highlighted as Region B). The potential cause of the development of these higher-temperature mixed-phase clouds in outbreaks are discussed in Section 6.4.

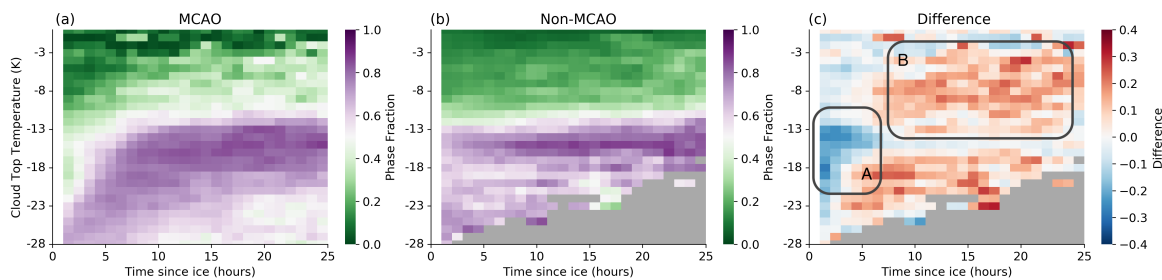


Figure 6.2: Evolution of mixed-phase cloud fractions as a function of time and cloud top temperature for (a) MCAO, (b) non-MCAO. (c) is the difference between them. Grid points with fewer than 500 successful retrievals are coloured in grey. In plot (c), red indicates that there are more mixed-phase clouds in MCAOs for that given cloud top temperature and TSI than in non-MCAOs, and blue indicates the opposite.

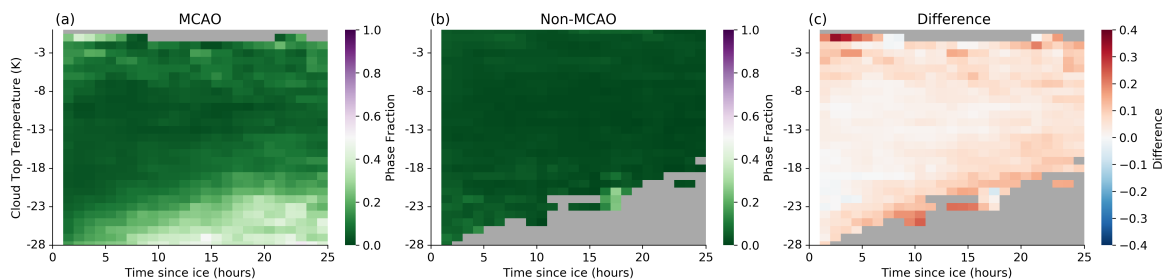


Figure 6.3: Evolution of ice phase fraction as a function of time and cloud top temperature for (a) MCAO, (b) non-MCAO. (c) is the difference between them. Grid points with fewer than 500 successful retrievals are coloured in grey.

6.3.3 Seasonal variability in mixed-phase development

Previous work has found a strong seasonal cycle in MCAO, with a maximum frequency of occurrence in the winter and minimum in the summer months (Fletcher et al., 2016b). Figure 6.4, which accounts for the MCAO index just as the air parcel leaves the ice edge, shows the number of MCAO pixels in the northern hemisphere summer in the region of study are orders of magnitude less common than other seasons for the period of study.

Figure 6.5 shows that for most seasons, the pattern of mixed-phase cloud development in MCAOs is similar to the annual aggregate; initially, mixed-phase clouds are only dominant at very low temperatures, and become increasingly dominant at higher temperatures until a plateau at around -13°C , although SON also has more

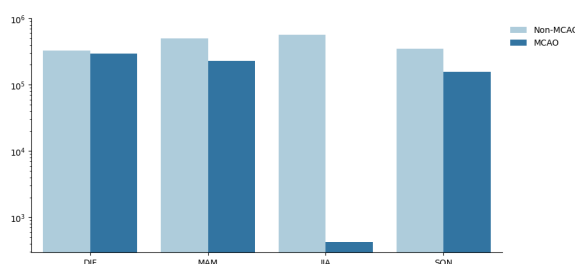


Figure 6.4: Number of MCAO and non-MCAO pixels recorded at $TSI = 1$ for each season.

mixed-phase clouds at higher temperatures later in the outbreak. JJA is an anomaly; its behaviour is very similar to the annual non-MCAO cloud development (Figure 6.2 (b)).

In contrast, Figure 6.6 shows the seasonal development of clouds in non-MCAO events. This time, MAM, SON and JJA replicate the annual pattern shown in Figure 6.2 (b). However, the non-MCAO DJF mixed-phase cloud development more strongly resembles an MCAO event (Figure 6.2 (a)). A potential cause for seasonal variation in both MCAO and non-MCAO mixed-phase development is discussed in Section 6.3.4.

6.3.4 Effects of air mass history

Initially focusing on the annually-averaged general cases, there are several possible causes of the distinctive mixed-phase evolution in the MCAO clouds seen in Region A of Figure 6.2 (c). One may be due to the air mass history. Figure 6.7 (a) shows the duration of time that air parcels which form MCAO and non-MCAO events typically spend moving over the sea ice before crossing over to open ocean. The pack ice is typically a poor source of INPs, with low INP concentrations being associated with longer times over ice (Porter et al., 2022; Li et al., 2023). Although AOD is an imperfect proxy for INP, especially in a region as poorly constrained as the Arc-

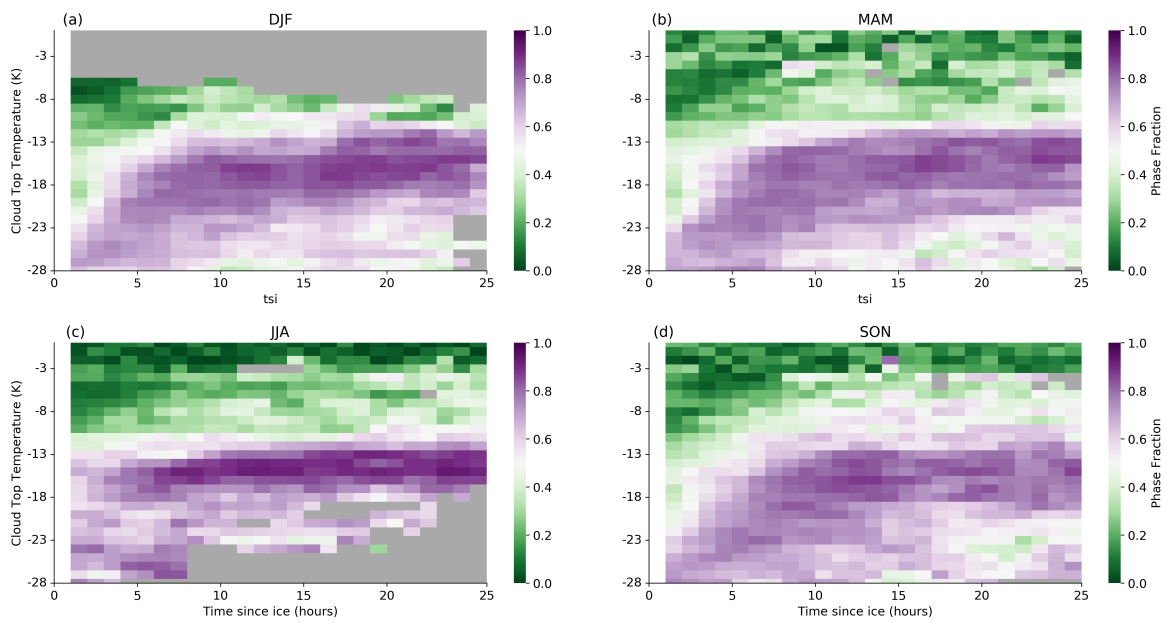


Figure 6.5: Evolution of cloud phase as a function of time and cloud-top temperature (from the DARDAR product) for MCAO clouds in (a) December, January and February, (b) March, April and May, (c) June, July and August and (d) September, October, November. Points for which there were fewer than 500 retrievals are coloured grey.

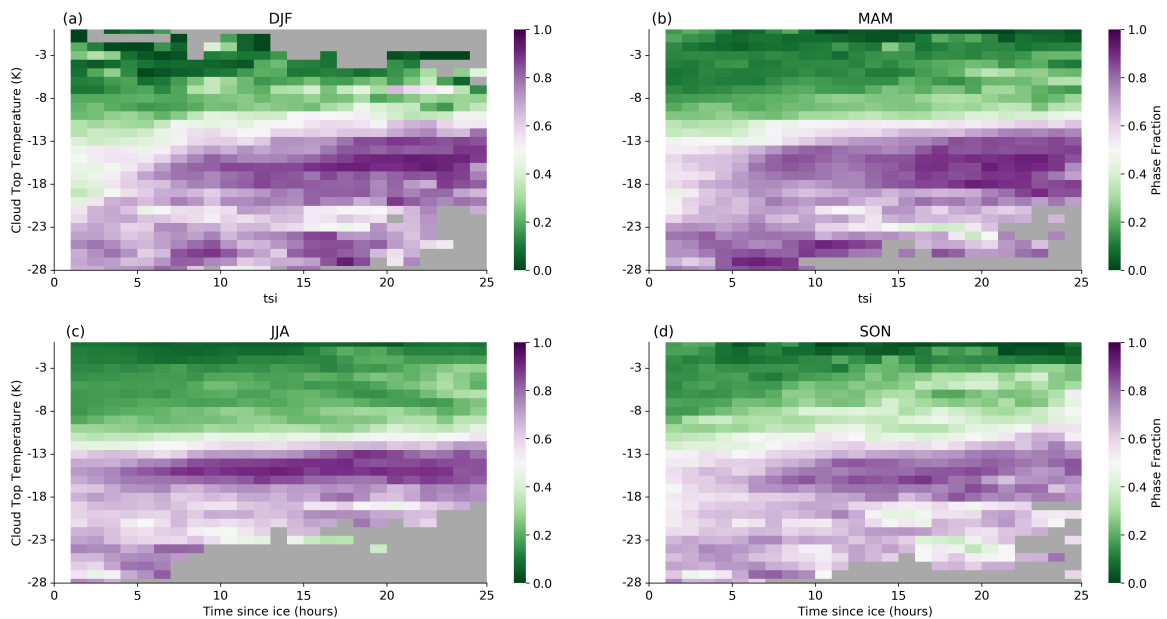


Figure 6.6: As Figure 6.5, but for non-MCAO clouds.

tic, Copernicus Atmosphere Monitoring Services AOD reanalysis data (Inness et al., 2019) was used to indicate the cleanliness of the air masses just as they move off of the ice edge (at a TSI = 1). Figure 6.7 (b) indicates that these MCAO air masses

typically have lower AODs than non-MCAO air masses, albeit the difference is small. Therefore, as they move off the ice edge, INP may be locally sourced from the ocean surface and transported to the cloud layer, and then activated to form ice particles. The higher wind speeds typically observed in MCAOs (Kolstad, 2017) may aid the transport of these INP via the bubble-bursting mechanism (Wilson et al., 2015), which has previously been observed (Inoue et al., 2021). The temperature of the liquid/mixed-phase switch (approximately -13°C after 7 hours in MCAO) is often considered the temperature at which biological INP tend to dominate over other potential INP, such as mineral dust (Murray et al., 2012). Therefore, it appears that while biological INP may already be present in the non-MCAO clouds as they move over the ice edge (indicated by the presence of mixed-phase clouds at -13°C close to the ice edge), they are absent from the MCAO air masses, and must be from a local source.

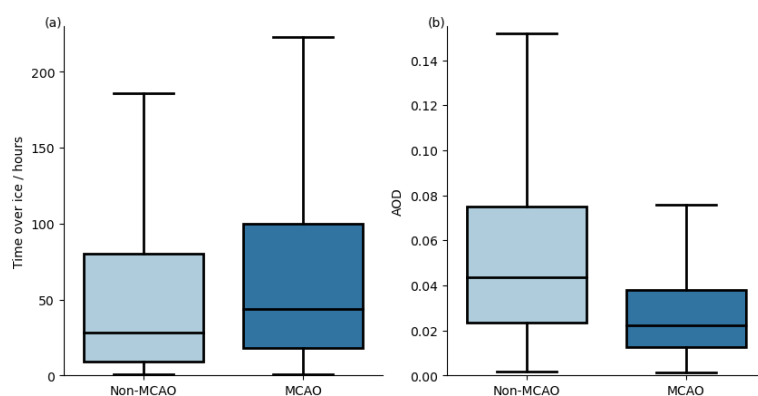


Figure 6.7: (a) Time spent over ice before leaving the ice edge and (b) AOD for MCAO and non-MCAO trajectories, just as the air leaves the ice edge (at TSI = 1).

Seasonal dependence of air mass history

What causes the non-MCAO DJF clouds to develop like most MCAO clouds and MCAO JJA clouds to develop like most non-MCAO clouds? One possible explanation is that the DJF non-MCAOs are very clean, therefore causing longer times for

the mixed-phase clouds to form. Similarly, one may expect the JJA MCAOs to have higher aerosol loads, therefore allowing the mixed-phase to rapidly develop. In this case, one would expect the time spent over ice to be important, due to the potentially cleaner air masses with longer times over ice. However, Figure 6.8 (a) shows that both DJF cases generally spend less time over ice than the other cases, contrary to what would be expected. Additionally, the JJA MCAOs spend comparable amounts of time over ice to MAM and SON cases (although, as there are fewer MCAO cases during the summer, this may be due to noise). While the CAMS AOD data (Figure 6.8 (b)) shows that DJF MCAO and non-MCAO are both relatively clean, the SON non-MCAOs and JJA MCAOs also have similar AOD. This may be due to CAMS not capturing the types of aerosols present in SON and JJA, which are expected to be dominated by local biological sources which are absent in the DJF case.

Looking further back into the air mass history provides some potential answers. Figure 6.9 shows the average time that the air parcels spent over snow-covered land before then moving onto ice for each season. Both non-MCAO and MCAO cases in winter typically spend long times over snow-covered land before reaching the ice, while the JJA cases typically spend very little, as expected when considering the seasonal cycle of surface cover (Figure 6.10). For both MAM and SON, the MCAOs usually spend a longer time over snow than non-MCAOs. Carlsen and David (2022) suggested that snow-covered surfaces prevented biological INP release and therefore suppressed mixed-phase formation at higher temperatures. Therefore, it may be that in summer, terrestrial INP are collected from the surrounding land mass and are available, along with local, marine biological INP, for ice formation. Terrestrial INP have previously been found to be an important source of higher-temperature INP in the summertime Arctic (Pereira Freitas et al., 2023). In contrast, in winter, no such INP are available for either the MCAO or non-MCAO events when the air is advected over land, and therefore it takes time for the local INP to be transported

from the ocean surface to the cloud layer in both cases. Although marine biological INP are scarcer in the winter, they are not entirely absent (e.g, Creamean et al. 2022; Hartmann et al. 2020; Pereira Freitas et al. 2023).

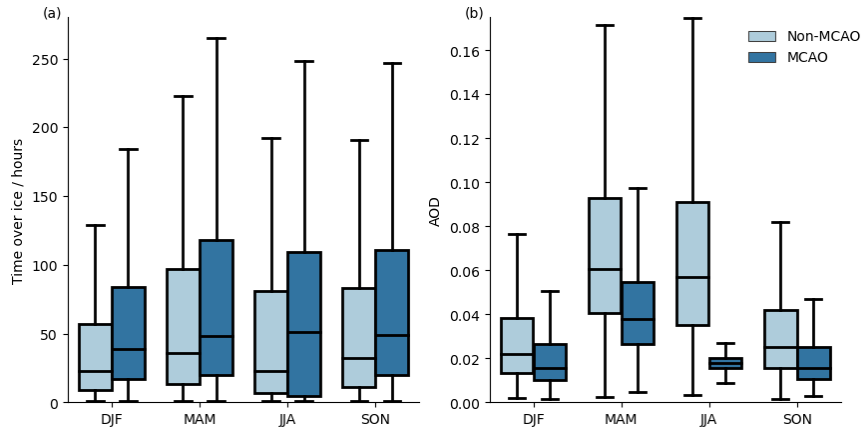


Figure 6.8: As Figure 6.7, but broken into seasons.

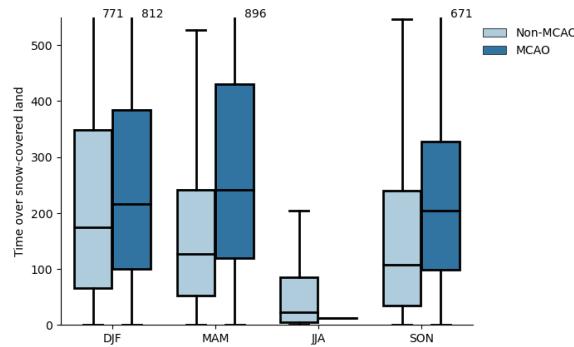


Figure 6.9: As Figure 6.8, but for time spent over snow-covered land before moving onto the ice edge.

To test the idea that the time spent over ice- and snow-covered surfaces impacts the temperature at which mixed-phase clouds initially form through INP availability, Figure 6.11 shows the development of MCAO clouds for DJF in cases where the air parcel has spent either more or less than two days over these INP-limiting surfaces. As is highlighted on Figure 6.11 (c) Region C, those cases which spend less time over sea ice form ice at higher temperatures within the first few hours over ocean. In contrast, the cases which have spent longer times over ice are typical of the gen-

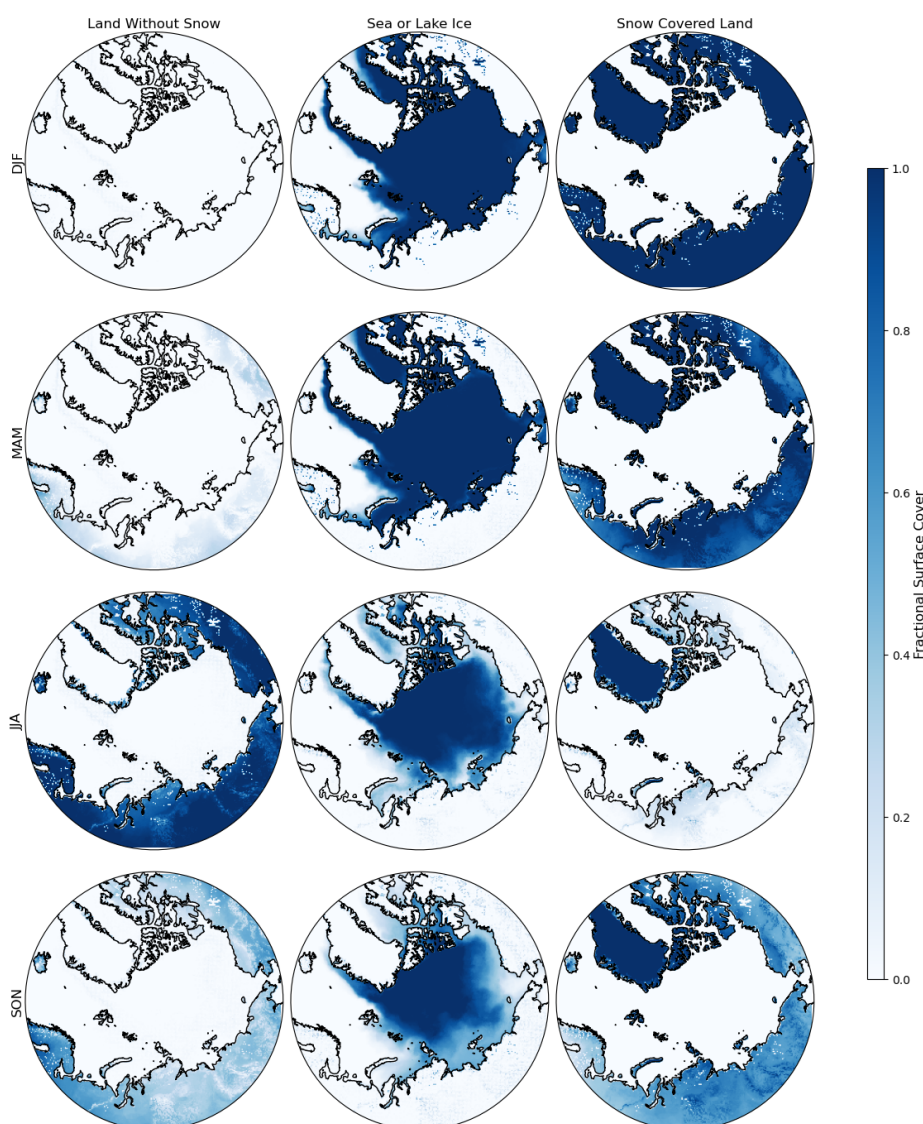


Figure 6.10: Seasonal climatologies of the fractional cover for different surface types; land without snow, sea or lake ice and snow covered land.

eral MCAO cases, suggesting that they are devoid of higher-temperature INP as they move off the ice edge and these aerosols must be sourced from the ocean surface. Additionally, the difference in phase development is pronounced only at the early stages of the trajectories. At later times and lower temperatures (Figure 6.11 (c) Region D), the phase compositions are very similar. This further suggests that air mass history primarily affects the formation of mixed-phase clouds, due to their reliance on INP availability.

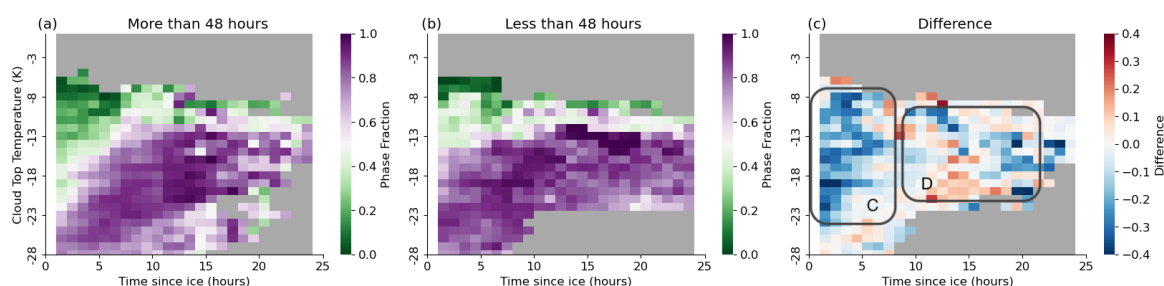


Figure 6.11: As Figure 6.2, but for DJF MCAO cases that have spent (a) more than 48 hours and (b) less than 48 hours over snow- and ice-covered surfaces. (c) is the difference between (a) and (b). Grid points with fewer than 250 successful retrievals are coloured in grey. Blue indicates that the cases which have spent longer times over ice have lower mixed-phase cloud fractions.

In summary, for the initial development of ice as the cloud moves from the ice edge, the air mass history appears to influence ice formation in both the MCAO and non-MCAO cases. MCAO air masses typically spend more time over ice- or snow-covered surfaces. As these surfaces may suppress the transport of INP to the atmosphere (e.g. Carlsen and David 2022), they may be relatively clean by the time they traverse the ice-ocean boundary. As the air moves over the ocean, the turbulent fluxes and strong wind allow for the transport of INP to the cloud layer, allowing marine INP to form ice. In contrast, non-MCAO typically spend less time over these INP-blocking surfaces, allowing for non-local INP to be carried over the ice edge, and thus earlier mixed-phase formation. There are two exceptions; in winter, the surrounding land is covered in snow and ice, so terrestrial INP aren't available, and there is only a weak source of marine biological INP. Additionally, the longer time spent travelling over these surfaces means that any existing INP in the air mass is more likely to be removed without replenishment. Therefore, the wintertime non-MCAO clouds develop very similarly to the MCAO clouds. However, if the DJF air masses spend relatively little time over ice, mixed-phase clouds do form at around -13°C close to the ice edge due to the availability of INP. In contrast, abundant terrestrial and biological INP in summer allow for the ready formation of ice in the MCAO. In spring and autumn, although non-MCAO clouds do travel over ice-covered surfaces, the bi-

ological sources are still active enough to allow for quick ice formation as the clouds move from the ice edge.

6.4 Potential causes of the higher-temperature mixed-phase clouds

The prevalence of mixed-phase clouds with CTT warmer than -10°C (Region B in 6.2 (c)) is challenging to explain with the data used here. There may be several causes of these higher-temperature mixed-phase clouds, namely: secondary ice production, the presence of INP active at these high temperatures and potential retrieval biases, which are discussed below.

6.4.1 Secondary ice production

Secondary ice production has previously been cited as a cause of mixed-phase development in MCAO; during a field campaign, Abel et al. (2017) found that the number of ice crystals in the post-transition cumuliform clouds was several orders of magnitudes greater than what would be expected given the ambient INP concentrations. Mages et al. (2023) also found evidence of secondary ice production in the later stages of an outbreak. In both cases, cloud temperatures were within those of the Hallett-Mossop mechanism and strong updrafts were recorded. The presence of moderate to strong updrafts has previously been observed to be important to the initiation of secondary ice production (Sullivan et al., 2018; Korolev et al., 2020; Luke et al., 2021). As non-MCAO clouds are not subject to the same instability as MCAO clouds, and therefore do not have as strong updrafts, this may explain the absence of the mixed-phase at higher temperatures. Although the mixed-phase enhancement

can be seen at lower temperatures than what is usual for the Hallett-Mossop temperature range (down to about -12°C in Figure 6.2 (c)), this may be due to the fact that the cloud top temperatures are used; the cloud base would be warmer than this, and therefore potentially in the appropriate temperature range. While temperatures up to -3°C may appear relatively warm for clouds associated with MCAOs, this may be due to the satellite observing convective clouds at different stages of development, as some clouds transition through these warmer temperatures until they reach their final cloud top temperature (Lensky and Rosenfeld, 2006). However, for secondary ice production to occur, there must be some initial primary ice nucleation; this may suggest that several mechanisms contribute to these higher-temperature mixed-phase clouds.

Secondary ice production is thought to be more efficient in the presence of larger liquid droplets (Rangno and Hobbs, 2001; Rosenfeld et al., 2011; Luke et al., 2021). To investigate this, the trajectories in Figure 6.2 (a) were divided into high and low r_e based on r_e retrievals taken within 6 hours of the DARDAR retrieval used in Figure 6.2. The r_e data were obtained from MODIS, and due to the uncertainties associated with retrieving r_e in mixed-phase clouds (Khanal and Wang, 2018), liquid-only retrievals were used. The aim is to consider the size of liquid droplets in the cloud before they transition to a mixed-phase regime. This excludes many cases for which a liquid-only MODIS retrieval could not be made (particularly during polar night, as MODIS requires reflected sunlight to retrieve r_e). A threshold of $14\text{ }\mu\text{m}$ was used to demarcate high and low r_e . The results are shown in Figure 6.12.

Due to the seasonal bias in MODIS retrievals, the general mixed-phase development in Figure 6.12 most closely resembles the JJA MCAO (which, in turn, resembles the non-MCAO clouds). Therefore, it is uncertain how representative these results are. However, it can be seen from Figure 6.12 (c) that clouds which had larger droplets

earlier in the development did go on to produce more mixed-phase clouds at the higher temperatures observed in Figure 6.2. The enhancement exists across the temporal development, although it is stronger after about 7 hours. Although not conclusive, this supports the argument that secondary ice production gives rise to these higher-temperature mixed-phase clouds. The reduction in mixed-phase clouds at lower temperatures for higher effective radius is due to an increase in ice-only clouds (not shown), as larger droplets also glaciate more readily (Coopman et al., 2020).

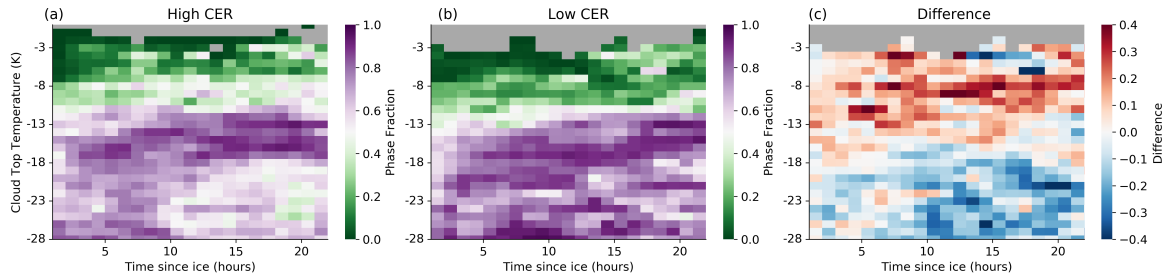


Figure 6.12: Evolution of mixed-phase cloud fraction as a function of time at cloud top temperature for clouds in MCAO events that have successful MODIS r_e retrievals for the liquid phase (a) above $14 \mu\text{m}$, (b) below $14 \mu\text{m}$. r_e retrievals must be at most 6 hours before the DARDAR retrieval to be included. (c) shows the difference between (a) and (b). Grid points with fewer than 250 successful retrievals are coloured in grey. Note that due to data limitations this threshold is lower than for Figure 6.2. Red indicates that clouds which have larger droplets earlier in the trajectory go on to have higher mixed-phase fractions than clouds which had smaller droplets.

6.4.2 Higher temperature INP

Another possible explanation is the activation of INP at these higher temperatures (between -10°C and -3°C). Biological INP in the seas around the ice pack have been recorded as being active as warm as -5°C (McCluskey et al., 2018; Irish et al., 2019; Wex et al., 2019; Li et al., 2023); albeit at low concentrations. Due to the scarcity of INP which would nucleate ice, it may take longer for them to activate, leading to the delay in the formation of these higher-temperature mixed-phase clouds. However,

as these higher-temperature INP are biological, a strong seasonal cycle is expected; although Figure 6.5 shows some seasonal differences, there is not a notable peak in summer, where biological activity is highest. Furthermore, it does not explain why this phenomenon is seen in MCAO but not non-MCAO clouds, unless the enhanced updrafts and instability in MCAOs greatly increased the transport of these INP to the cloud layer, evidence of which has been observed previously (Inoue et al., 2021). More measurements of INP around MCAO would be required to determine the contribution of high-temperature biological INP to mixed-phase cloud formation. Specifically, measuring the prevalence of biological INPs across different seasons, how their abundance changes with distance from the ice edge and examining how their ice-nucleating capabilities vary with temperature would help to interrogate the hypotheses presented in this study.

6.4.3 Retrieval biases

It is possible that the higher-temperature mixed-phase clouds are not a real effect but appear due to the radar misclassifying supercooled drizzle droplets as ice, particularly above -10°C , due to their larger size (Zhang et al., 2017, 2018). As noted by Danker et al. (2022), this may be a particular issue with DARDAR when the lidar is attenuated in supercooled-topped clouds, and heavier precipitation would be required for a CloudSat detection (Marchand et al., 2008). Drizzle has previously been observed to form in MCAOs (Abel et al., 2017; Lloyd et al., 2018), and supercooled drizzle in general has been recorded at temperatures down to -25°C (Silber et al., 2019). Additionally, droplets in liquid clouds formed during MCAOs grow to precipitation-size (about $15\text{ }\mu\text{m}$; Rosenfeld and Gutman 1994), whereas those not in outbreaks do not (Chapter 5), potentially explaining the differences between Figure 6.2 (a) and (b). Schirmacher et al. (2023) found that CloudSat may also underesti-

mate precipitation, particularly frozen precipitation, in outbreaks due to limitations such as surface clutter creating a blind zone for the radar close to the surface. While the COMBLE campaign took in situ precipitation measurements during outbreaks, and found that the cumuliform clouds were typically precipitating (Mages et al., 2023), with frozen precipitation being common, there is still a great deal of uncertainty about the phase of the precipitation created in these events. Therefore, it is difficult to assess how prevalent retrieval errors may be. More in situ measurements of the frequency and characteristics of supercooled drizzle would be required for a more robust understanding of precipitation in these events and to target the retrieval uncertainty.

With the data used in this work, identifying the cause of mixed-phase clouds forming above -10°C late in the MCAO events is challenging. Some evidence such as the influence of droplet size and the absence of similar formations in non-MCAO events suggests secondary ice production, which is consistent with in situ observations of MCAO clouds. However, I cannot conclusively rule out other potential causes of the appearance of Region B in Figure 6.2. Other factors, such as snow lofted from the surface or from below the cloud layer nucleating ice in the cloud (Geerts et al., 2015), cannot be ruled out with the current analysis. This may be especially relevant in MCAO due to the high wind speeds, turbulence and the presence of frozen precipitation (Mateling et al., 2023). Satellite data alone is unlikely to be able to determine the formation mechanism of these late-stage mixed-phase clouds, highlighting the need for more detailed observations of this phenomenon. Current and upcoming measurement campaigns, such as the Cold Air Outbreak Experiment in the Sub-Arctic Region (CAESAR) campaign, would be well suited to target this uncertainty (National Center for Atmospheric Research, 2024).

6.5 Discussion

The results presented here are consistent with the air mass history influencing the development of mixed-phase clouds, both in MCAO and non-MCAO events. However, this trajectory analysis does not allow for the precise origin of the air masses to be determined, and therefore there are uncertainties in linking these trajectories to potential INP sources in the wider Arctic region. Additionally, there may be potential INP sources in the pack ice, such as open leads, which have previously been cited as aerosol sources at high latitudes (Bigg and Leck, 2001). These features may be on the order of several hundred meters in size (Wadhams et al., 1985; Li et al., 2020), and therefore below the resolution of the sea ice product (Cavalieri et al., 1996). However, Porter et al. (2022) found that open leads were very weak sources of INP, and that air that had spent time over the pack ice typically had very low aerosol concentrations. Hartmann et al. (2020) found that INP from leads were active at high temperatures and suggested that they were from a biogenic source. Therefore, if INP from features like open leads were prevalent in MCAOs, one may expect more mixed-phase cloud formation at higher temperatures as soon as the air moves off the ice edge (low TSI) in Figure 6.2 (a). As open leads cover a higher fraction of the sea ice area in summer than winter (Lindsay and Rothrock, 1995; Wang et al., 2016), these features may be partially responsible for the similarity between the MCAO and non-MCAO JJA cases. However, there is also a high fractional coverage of leads in the autumn, and yet the MCAO and non-MCAO development remain distinct. As there is disagreement regarding the efficacy of leads acting as INP sources, more measurements would be required to determine their influence on cloud phase processes in the Arctic region. Whether it be through sources such as leads or INP transported from adjacent snow-free land, these results still indicate the importance of air mass history in the formation of the mixed-phase clouds.

It should also be noted that the analysis here is based on the cloud top temperature, and the DARDAR data product derives cloud top temperature from ECMWF-AUX (Partain, 2022). This may introduce uncertainties. Furthermore, the DARDAR phase classification algorithm also uses the ECMWF wet bulb temperature to distinguish between ice (below 0°C) and liquid or rain (above 0°C), which may reduce the phase classification accuracy at higher temperatures (such the mixed-phase clouds seen in Figure 6.2 (a)). However, previous work has found good agreement between the cloud top temperature when using ECMWF-AUX temperatures and in situ radiosonde data (e.g., McErlich et al., 2021) or other retrieved satellite data (e.g., Christensen et al., 2016), so the effects of potential biases for these may be limited.

Mixed-phase clouds are of interest due to the effect of ice on the lifetime of the stratocumulus clouds, with higher INP concentrations hastening the breakdown of the high-albedo stratocumulus deck through precipitation-related mechanisms (Vergara-Temprado et al., 2018; Murray et al., 2021; Tornow et al., 2021). However, due to limitations with CloudSat in MCAOs (Schirmacher et al., 2023), DARDAR struggles to detect precipitation in these events. Therefore, I am unable to determine the effects of the formation of ice on the precipitation frequency in these clouds. Additionally, MODIS retrievals struggle with mixed-phase clouds, which are often topped by a supercooled liquid layer and therefore may be retrieved as liquid-only clouds. An analysis on the effects of ice on cloud properties and lifetime is also beyond the scope of this research. To achieve this, high-resolution measurements, both spatially and temporally, of ice formation and the effects on properties such as cloud fraction and water path would be required.

6.6 Conclusions and Outlook

Marine cold-air outbreaks clouds undergo a distinctive evolution from high coverage stratiform decks to broken cumuliform cloud fields. These post-transition clouds typically have a much lower shortwave cooling effect due to their lower cloud fractions. Therefore, factors which may change the timing of the transition can change the role these clouds play in the Arctic energy budget. In particular, the presence of ice or frozen precipitation is thought to be key to the cloud evolution. However, the conditions influencing ice formation in these events are uncertain. Here, I have developed a method to observe the evolution of the cloud phase over time in these outbreaks, and considered the role of air mass history on the cloud evolution.

After leaving the ice edge, MCAO clouds exhibit a rapid decline in the liquid phase, with a corresponding increase in mixed-phase clouds, and some amount of glaciation into ice-only clouds (Figure 6.1). In contrast, the liquid phase is persistently dominant in non-MCAO clouds, with particularly few ice-only clouds. The shift towards a more glaciated state in MCAO clouds is in agreement with previous in situ and measurement campaign observations (Abel et al., 2017; Lloyd et al., 2018; Mages et al., 2023), with higher amounts of ice found in the post-transition cumuliform clouds than in the original stratiform decks.

Mixed-phase clouds predominantly exist at low temperatures initially (around -20°C), gradually shifting to form at higher temperatures of around -13°C (Figure 6.2). In contrast, mixed-phase clouds form at higher temperatures in non-MCAO conditions very close to the ice edge. There is a seasonal dependence to the formation of ice in MCAOs and non-MCAOs (Figures 6.5 and 6.6), with wintertime non-MCAO clouds only forming mixed-phase at low temperatures initially and JJA MCAO forming ice at -13°C at the ice edge. These differences in evolution appear to

be related to air mass history; air masses which form MCAO typically spend longer times over ice- and snow-covered surfaces, which usually are poor sources of INP (Figures 6.8 and 6.9). Therefore, the time taken to form mixed-phase clouds at higher temperatures appears to be associated to the transport of local, marine biological INP to the cloud layer. In contrast, non-MCAO events typically spend less time over snow and ice, and therefore may carry more INP. Although the CAMS AOD data somewhat corroborates the results, with MCAO air parcels typically being cleaner, the difficulties in representing biological sources and INP in models limits the utility of the reanalysis data.

Despite several measurement campaigns, there is still considerable uncertainty about aerosol sources and sinks in the Arctic (Schmale et al., 2021). The results presented here highlight the need for more accurate measurements of the sources of biological INP in the region, particularly close to the ice edge, and the mechanisms of transport to the cloud layer. Further knowledge of aerosol transportation pathways around the Arctic is also required, especially for understanding the seasonal differences in aerosol sources.

Further into the MCAO development, there is some evidence of mixed-phase clouds occurring with cloud top temperatures above -10°C (Figure 6.2). Although evidence of secondary ice production has previously been observed in MCAO clouds, particularly in regions of strong updrafts or high supercooled fraction (Abel et al., 2017; Mages et al., 2023), it is difficult to determine decisively with satellite data if the mechanism is at play here. Karalis et al. (2022) showed that secondary ice formation was important for the development and break-up of MCAO clouds, particularly mechanisms such as drop-shattering and ice-ice collisional break-up. However, parameterisations of these mechanisms are highly uncertain and not widely implemented in climate models. Further laboratory and in situ studies are required to

understand these mechanisms, which can improve the representation in the models and in turn help to understand whether or not secondary ice production contributes to the higher-temperature mixed-phase clouds observed in MCAO.

This study provides evidence consistent with previous modelling work showing that MCAO cloud development is sensitive to INP concentration (Vergara-Temprado et al., 2018). Future changes in Arctic INP concentrations are expected to change the radiative properties of the clouds. Increasing temperatures mean that the Arctic is increasingly snow- and ice-free, therefore potentially making more INP sources more available for greater parts of the year (Ardyna et al., 2014). These increases in INP may further increase the proportion of mixed-phase clouds in MCAOs, or help ice form earlier into the outbreak, and cause these clouds to break up more rapidly (Tornow et al., 2021). During polar day, both of these changes would decrease the cooling effects of the clouds, leading to a positive feedback - however, the strength and even sign of this feedback may be sensitive to changing strength and occurrence of MCAO in a future Arctic. Additionally, poor parameterisations of INP in climate models, along with difficulties of modelling mixed-phase cloud properties and the dynamics of a complicated system like a MCAO, mean that understanding the impact of changes in aerosol concentrations is challenging. To accurately estimate the climate impact of this effect on a large scale, it is necessary to conduct detailed measurements of both aerosols and mixed-phase clouds across the Arctic.

Chapter 7

Conclusions

Clouds are central components of the global energy budget. Aerosols can strongly influence the radiative effect of clouds through the modification of their properties, but these interactions are the largest uncertainty in global climate projections. This issue is particularly salient in the Arctic, a region already undergoing significant transformation due to anthropogenic climate change. Shifts in the surface energy budget induced by Arctic clouds have been linked to variations in sea ice extent (Kay and Gettelman, 2009; Huang et al., 2019) and melting of the Greenland ice sheet (Bennartz et al., 2013). With ongoing changes such as increased industrialisation and rapid surface warming, understanding the aerosol-cloud relationship, in particular for radiatively important properties CF and LWP, is vital to resolve what these changes mean for the region at large.

Arctic cloud properties have been the subject of several in situ measurement campaigns over recent decades (e.g., Lin et al., 2003; Verlinde et al., 2007; Lloyd et al., 2015; Shupe et al., 2022), as well as being observed by long-running Arctic research stations such as that at Summit, Greenland (Shupe et al., 2013). However, due to the difficult operating conditions in the harsh Arctic environment, these campaigns are typically limited in both space and time. While these measurements offer invaluable

able insights into cloud properties, there are significant portions of the Arctic which have not been covered by these measurement efforts, in particular over the Arctic ocean. Therefore, it is difficult to determine how representative these measurements are over the whole region. Additionally, measurements from established sites are made in an Eulerian framework, and do not account for the dynamic changes in cloud properties and have limited spatial coverage. While flight campaigns may go some way into looking at timescales, they observe a limited number of samples (e.g., Young et al., 2016; Abel et al., 2017) and therefore it is difficult to test how different environmental conditions affect the cloud evolution. However, work in this thesis has shown that these conditions can strongly influence cloud properties downwind of the initial cloud formation (Chapter 5).

Previous studies which have considered the aerosol-cloud interactions have typically not considered the high latitudes due to known issues with the MODIS retrieval algorithm (e.g., Quaas et al., 2006; Michibata et al., 2016; Gryspeerd et al., 2019a). Those studies which use satellites in the Arctic tend to use active satellites, but this limits the cloud properties obtainable (for example, N_d cannot be derived from CALIOP data). Therefore, these works rely on reanalysis or model aerosol estimates to diagnose the relationship, despite the uncertainties with these products (Zamora et al., 2017; Coopman et al., 2018a). Similarly, previous research on cloud development has focused on lower latitudes to make use of frequent observations from geostationary satellites (Christensen et al., 2020), despite the fact that it is difficult to get N_d or LWP from these satellites (Smalley and Lebsock, 2023). However, the Arctic presents a distinct advantage for cloud evolution studies due to the high frequency of satellite overpasses from polar-orbiting instruments. Furthermore, although the development of mixed-phase clouds, especially in the Arctic, has been identified as a critical research area (de Boer et al., 2022), phase transitions have received limited attention in previous cloud development studies.

This thesis has shown that using satellite data can overcome some of the difficulties encountered by in situ studies. In particular, the state-of-the-art cloud property data from MODIS can significantly enhance our understanding of Arctic clouds. Chapter 3 showed that while using these data in the Arctic can be difficult, namely in the cases of low sun and misclassified ice, careful filtration of the MODIS data can reduce bias, creating a reliable dataset which can be used to probe the impact of aerosols on Arctic clouds.

Using MODIS data, Chapter 4 considered the relationship between LWP - a key determinant of cloud albedo - and aerosols, using N_d as a mediating variable. This analysis revealed significant geographical variations in the N_d -LWP sensitivity. In particular, a positive relationship was observed at higher latitudes, in contrast to the typically negative correlation observed in previous global satellite studies. Lower tropospheric stability was found to be the most significant control on the sign of this relationship. Further investigation into the non-linearities of the relationship revealed a temperature-dependent aerosol effect, as sensitivity of LWP to aerosol changed with LTS, which is predominantly governed by the surface temperature in the Arctic. This finding is particularly significant given that many studies on aerosol-cloud interactions conventionally assume that the nature of the relationship is independent of the surface temperature, and therefore may not capture potential future changes in radiative forcing with surface warming. This work also suggests an aerosol-dependent cloud feedback, as the LWP-LTS (and therefore surface temperature) relationship also depends on the aerosol state. In the context of a warming, more polluted Arctic, this suggests a diminished cooling effect from aerosol adjustments, which would need to be accounted for when considering the future Arctic energy budget.

Chapter 5 expanded on the use of snapshot satellite images in Chapter 4 by developing a method to characterise the temporal development of clouds formed during marine cold air outbreaks. This analysis provided novel insights into the lifecycle of MCAO clouds, observing the transition from stratiform to cumuliform fields. In particular, cloud fields which formed in regions with higher aerosol concentrations typically broke down up to 7 hours later than those which formed in cleaner conditions, demonstrating an aerosol effect on cloud lifetime. This was due to suppressing droplet growth and delaying precipitation, which was found to be key to the transition to cumulus clouds. The cloud fraction was found to be the leading control on determining cloud albedo and consequently the effect of these clouds on the surface energy budget. This work extends our understanding of cloud dynamics over the Arctic Ocean and highlights the necessity of considering temporal evolution in fully understanding effects of aerosols on cloud properties.

While Chapter 5 considered liquid-dominated clouds, Chapter 6 advanced this analysis to explore the effects of air mass history on the formation of mixed-phase clouds in the Arctic. Clouds in MCAO rapidly shift to ice-containing states as the air moves off the ice edge, in contrast to non-MCAO clouds which maintain a steady mixed-phase fraction. These mixed-phase clouds in MCAOs initially form at lower temperatures (below -20°C) and later are predominant at around -13°C , a temperature usually associated with the activation of biological INP. This work revealed the crucial role of air mass trajectories, particularly over ice- and snow-covered surfaces, in determining the formation of ice in these clouds due to the scarcity of INP over these surfaces. There is also some evidence for secondary ice production mechanisms forming mixed-phase clouds as high as -5°C late into the trajectory. This provides a unique perspective on the factors affecting temporal development of mixed-phase clouds and highlights the need for understanding INP sources and sinks in the Arctic to accurately capture cloud formation and processes.

7.1 Suggestions for Future Research

Ice processes and cloud break up

Chapter 5 explores the influence of precipitation on the break up of liquid-dominated clouds. Modelling studies suggest that ice hydrometeors play a crucial role in determining when these mixed-phase clouds transition (Tornow et al., 2021); however, Chapter 6 could not link the formation of ice to precipitation dynamics due to the limited coverage of active sensors, which restricts observations to a single pass over each MCAO event. Addressing this gap requires additional satellite missions and focused measurement campaigns to analyse ice concentrations before and after precipitation events, enhancing our understanding of cloud phase transitions. Furthermore, such campaigns could investigate the role of secondary ice production mechanisms, which are highly uncertain and have been identified as a significant area for further research in understanding Arctic cloud dynamics (de Boer et al., 2022). The European Space Agency’s Earth Cloud Aerosol and Radiation Explorer (EarthCARE), due to launch in May 2024, carries a sensitive cloud profiling radar and promises to provide detailed measurements of particle size distribution, cloud water content and hydrometeor vertical velocity. Integrating these data with pseudo-Lagrangian trajectory analysis promises to shed light on the impact of ice microphysics, including secondary ice production, on the dissipation of clouds.

Constraining cloud processes in models

One of the primary motivations for this thesis is to constrain the impacts of aerosols on clouds such that they are better represented in climate models. This is particularly important for the Arctic, as cloud physics parameterisations are often developed for lower latitudes and are assumed to be suitable for the Arctic (Kay et al., 2016). As

observations are crucial for model evaluation, this assumption may have been necessary due to the scarcity of observations of Arctic clouds. This research could help bridge this observational gap. Specifically, it offers insights into the timescales of cloud processes, identifying areas where model parameterisation schemes could be refined at the process level, which is key to then improving accuracy and reliability in capturing Arctic cloud properties. For example, the framework of investigating the temporal evolution of clouds presented here could be used to measure the timescales of precipitation formation or suppression and the relationship to aerosols. These results could be compared to the model process rates and potentially identify areas of further development. This could lead to better representation of cloud lifetimes and liquid water paths, which are two key areas where model improvement is needed.

Models can be used to assess the radiative impact of aerosol-cloud interactions identified in this thesis on the Arctic climate. Although this work provides estimates of the forcing due to aerosol-induced changes in cloud properties, models offer a pathway to more precisely quantify these effects. For example, models can examine the extent to which aerosol-cloud interactions influence Arctic sea ice extent and thickness. However, the variability in how aerosol-cloud interactions are represented across CMIP6 models has been recognized as a major obstacle in understanding the sea ice-cloud relationship (Luo et al., 2023), highlighting the need to first address model deficiencies.

Measurements of Arctic aerosol

The uncertainties surrounding Arctic aerosol sources, sinks, composition, and transport, particularly the role of INPs, present substantial challenges to accurately modelling clouds and understanding cloud-phase climate feedback mechanisms (Schmale et al., 2021; Murray et al., 2021). A key limitation of this thesis is its reliance on reanalysis data for estimating aerosol conditions, due to the sparse satellite coverage

in the Arctic and inherent uncertainties in reanalysis products. Targeted measurement campaigns could provide detailed, high-quality insights into cloud properties and atmospheric conditions, which is a key advantage over satellite-based studies. These data could be used to test hypotheses presented here. For example, although evidence presented in Chapter 6 suggests a significant impact of biological INPs on mixed-phase cloud formation, direct aerosol measurements within MCAO air masses would be needed to better constrain the relationship.

In particular, this research complements the work of recent and upcoming measurement campaigns, such as the HALO-(AC)³ mission (Wendisch et al., 2023), part of which is dedicated to advancing our understanding of Arctic air mass transformations within a pseudo-Lagrangian framework. Consideration of initial atmospheric conditions has typically been overlooked in previous measurement campaigns of MCAO clouds. However, the work here shows that these initial conditions should be considered as key targets for the campaign to improve our understanding of the cloud dynamics.

Moreover, extensive in situ sampling could provide more accurate insights into how variations in initial conditions influence cloud processes and the impact of aerosols on cloud transitions. As the work in this thesis also suggests a role for air mass history and long-range aerosol transport, integrating knowledge from existing field campaigns and measurement stations, along with targeting future missions towards known blind spots, would help to create a pan-Arctic picture of aerosol properties and better inform model and satellite studies.

Understanding the efficacy of marine cloud brightening

The urgency to combat Arctic sea ice loss due to rapid regional warming has prompted groups such as the Refreeze the Arctic Foundation (Refreeze The Arctic Foundation,

2023) to advocate for the use of marine cloud brightening (MCB). While the practical and moral considerations of MCB are beyond the scope of this thesis, the findings of Chapter 4 are pertinent to these efforts. MCB, which involves the artificial enhancement of cloud albedo through aerosol seeding to reduce solar radiation absorption by the ocean and limit sea ice melt, fundamentally relies on the Twomey effect (Latham et al., 2014). This research shows that LWP adjustments to aerosol are also vital to consider: in unstable environments, addition of aerosol could actually significantly offset the Twomey effect, making the MCB efforts substantially less effective. Therefore, the strategic deployment of aerosol seeding vessels would require real-time measurements of LTS (among other meteorological variables). While successful MCB could induce a feedback mechanism - cooler surface temperatures lead to more stable conditions, creating regimes in which adjustments enhance the Twomey effect - a better understanding of both precipitation and entrainment mechanisms is needed to project LWP adjustments in different environmental conditions (Diamond et al., 2022).

Appendix

A.1 The calculation of the non-precipitating AMSR-E LWP

The additional cloud liquid water needed to convert the AMSR-E LWP product above 180 g m^{-2} to the non-raining LWP is based on the assumption that the rain water content (RWC) can be calculated using:

$$RWC = \frac{4\pi}{3} \rho \int r^3 n(r) dr \quad (\text{A.1})$$

$$n(r) = N_o e^{-\Lambda r} \quad (\text{A.2})$$

$$\Lambda = CR^{-0.21} \quad (\text{A.3})$$

in which r is the droplet radius and $n(r)$ is the Marshall-Palmer droplet size distribution, and C and N_o are parameters derived from that distribution (Marshall and Palmer, 1948). R is the rain rate. Assuming that the RWC is constant across the raining column, Equation A.4 can be derived:

$$eLWP = \frac{8HN_o\rho\pi}{(CR^{-0.21})^4} \quad (\text{A.4})$$

using Equations A.5 and A.6 (Wentz and Spencer, 1998; Hilburn and Wentz, 2008):

$$H = 0.46 + 0.16T_{sst} \quad (\text{A.5})$$

$$R = \frac{(\frac{LWP}{\alpha} - 1)^2}{H} \quad (\text{A.6})$$

in which eLWP is the “extra” LWP above 180 g m^{-2} that would be added if the cloud were assumed to be non-raining, H is the height of the raining column, α is the AMSR-E rain threshold, ρ is the density of water, LWP is the cloud liquid water path derived by AMSR-E and T_{sst} is the sea surface temperature.

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