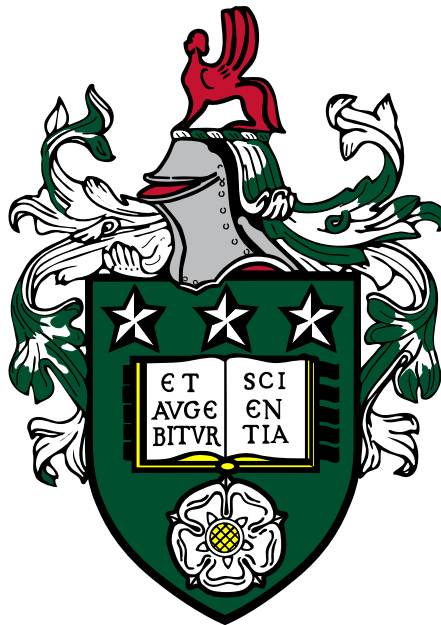


High-latitude ice-nucleating particles and the effects on mixed-phase clouds



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The candidate confirms that the work submitted is their own, except where work which has formed part of jointly authored publications has been included. The contribution of the candidate and the other authors to this work has been explicitly indicated below. The candidate confirms that appropriate credit has been given within the thesis where reference has been made to the work of others.

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David, R. O. and Carlsen, T. of the University of Oslo performed measurements of INPs at Andenes used in Chapter 4 and Chapter 5.

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Abstract

High-latitude mixed-phase cloud systems are important to the climate as their reflectivity will increase as the climate warms, since ice in the clouds will be replaced by liquid. This mechanism is called the cloud-phase feedback. Ice-nucleating particles (INPs) play a key role in these clouds, underpinning the amount of ice formation and modulating the strength of this feedback. However, there is limited understanding of the concentrations and sources of INPs in high-latitude mixed-phase clouds, or how INPs are processed by these cloud systems. To address these challenges, this thesis presents measurements and computer modelling of INPs in cold-air outbreaks (CAOs). These high-latitude mixed-phase cloud systems form a useful natural laboratory since they have predictable formation locations and well-defined structures. This work found that springtime CAOs over the Norwegian and Barents seas had some of the highest INP concentrations observed in the Arctic. These high concentrations were attributed to aged fertile dust aerosol in Arctic haze transported from lower-latitude regions. These observations also suggested that the INP concentration reduced through a CAO as INPs were removed through precipitation. To investigate the magnitude of this change, CASIM, a double-moment microphysics module with interactive aerosol was used to simulate a CAO. This modelling confirmed that INPs are removed through a CAO but the simulated rate of removal was less than measured. Though INPs were removed, there was a negligible impact on INP concentrations in the cloud, which were predominantly controlled by temperature. However, the ice nucleation scheme in CASIM did not track INP sites, instead assuming there was a constant relationship between the number of INPs and the aerosol concentration. Using a new ice nucleation scheme that tracked active sites, we were able to replicate the observed reduction of INP concentrations, but not while accurately simulating the magnitude of the INP concentrations.

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List of abbreviations

2-DS	2-Dimensional Stereo probe
ACAO	Arctic Cold-Air Outbreak campaign
ACCACIA	Aerosol-Cloud Coupling And Climate Interactions in the Arctic
ACTIVATE	Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment
ARCL	The aerosol climatology of Bellouin et al. (2013)
ARKTIS	A series of twentieth-century Arctic field campaigns.
CAO	[marine] cold-air outbreak
CAMS	Copernicus Atmosphere Monitoring Service
CAESAR	Cold-Air outbrEaks over the Sub-Arctic Region
CASIM	Cloud AeroSol Interacting Microphysics
CERES	Clouds and the Earth's Radiant Energy System (a satellite radiometer)
CCN	Cloud condensation nucleus/nuclei
CDP	Cloud Droplet Probe
CESM2	Community Earth System Model 2
CFDC	continuous-flow diffusion chamber
COMBLE	Cold-air Outbreaks in the Marine Boundary Layer Experiment
CRE	cloud radiative effect
Cu	cumulus
cXXX	Flight with identification number cXXX during the ACAO campaign
cXXXrY	INP sample with identification number Y taken during flight cXXX
DRINCZ	DRoplet Ice Nuclei Counter Zurich
EDS	energy-dispersive X-ray spectroscopy
ERA5	European Centre of Medium-range Weather Forecasts Reanalysis v.5
ERF	Effective Radiative Forcing

FAAM	Facility for Airborne Atmospheric Measurements
GDAS	Global Data Assimilation System
GPS	Global Positioning System
HALO-(AC)³	Project on Arctic Amplification Climate Relevant Atmospheric and Surface Processes and Feedback Mechanisms
HEPA	high efficiency particulate air [filter]
HM	Hallett-Mossop (process of secondary ice production)
HYSPLIT	HYbrid Single-Particle Lagrangian Integrated Trajectory
IGP	Iceland Greenland Seas Project
INP	ice-nucleating particle
IQR	interquartile range
IWC	ice water content (a measure of ice mass per unit volume of air, not to be confused with the ice mixing ratio, a measure of ice mass per unit mass of air)
lat.	latitude
lon.	longitude
LWC	liquid water content (a measure of liquid mass per unit volume of air, not to be confused with the liquid mixing ratio, a measure of liquid mass per unit mass of air)
MASIE	Multisensor Analyzed Sea Ice Extent
MIROC6	Model for Interdisciplinary Research on Climate, version 6
MODIS	MODerate resolution Imaging Spectrometer
MONC	Met Office–NERC Cloud Model
M-Phase	Resolving climate sensitivity associated with shallow mixed-phase cloud in the oceanic mid- to high-latitudes (the sister research project to ACAO)
MPOF	Mixed-phase overlap fraction
MURK	Aerosol coupling in the UKV model
N680	Global model resolution of $2N \times 1.5N$ grid cells
NERC	Natural Environment Research Council
NORESM2	NORwegian Earth System Model version 2
OSTIA	Operational Sea surface Temperature and Ice Analysis
PCASP	Passive Cavity Aerosol Spectrometer Probe
PDF	probability density function
PTFE	Polytetrafluoroethylene
RAL	Regional Atmosphere and Land

Sc	stratocumulus
SCT	stratocumulus-to-cumulus transition
SDD	silicon drift detector
SEM	scanning electron microscopy
SIP	secondary ice production
SOCRATES	Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study
UKCA	United Kingdom Chemistry and Aerosols model
UKV	The operational forecasting model of the UK Met Office
UM	UK Met Office Unified Model
UTC	Coordinated Universal Time
WBF	Wegener-Bergeron-Findeisen process

List of symbols

g	Graupel (as a model hydrometeor species).
i	Ice crystals (as a model hydrometeor species). Note this refers to the species only, and not to the total frozen water content, which would include snow and graupel.
r	Rain (as a model hydrometeor species).
s	Snow (as a model hydrometeor species).
v	Water vapour (as a traced model species).
w	Cloud water (as a model hydrometeor species). Note this refers to the species only, and not to the total liquid water content, which would include rain.
$a, b, \dots$	Arbitrary constants used in parameterisations. Values are defined at source with respect to their parameterisations.
$\bar{a}, \bar{b}, \dots$	Arbitrary constants used in parameterisations of average values. Values are defined at source with respect to their parameterisations.
a_{crit}	Critical radius of an ice embryo to nucleate.
a_i	Radius of an ice embryo (in the process of nucleation).
A_d	Surface area of filter in contact with a droplet in a droplet-freezing assay.
A_f	Surface area of filter exposed to aerosol in a droplet-freezing assay.
A_{sample}	The surface area of filter sampled during SEM.
C	Distance between two INP measurements
D	Aerosol diameter.
e_i	Saturation vapour pressure over ice.
e_w	Saturation vapour pressure over water.
$f(), g() \dots$	Arbitrary functions.
f_f	Fraction of droplets frozen in a droplet-freezing assay
F_{Cooper}	Prefactor in Cooper INP parameterisation (Cooper, 1986).
F_{aer}	Factor of change in total measured surface area up/downwind (see Section 2.3.5)
F_{INP}	Factor of change in INP concentration up/downwind (see Section 2.3.5)
F_S	Factor of change in INP concentration normalised by aerosol surface area up/downwind (see Section 2.3.5)
ΔG	Gibbs free energy change associated with ice nucleation.
ΔG^*	Energy barrier to ice germ formation in homogeneous nucleation.
ΔG_{het}^*	Energy barrier to ice germ formation in heterogeneous nucleation.

H	Cold air outbreak depth.
J_{hom}	Homogeneous nucleation rate according to classical nucleation theory.
k	Differential nucleus spectrum.
k_{assay}	Differential INP concentration for a droplet-freezing assay.
k_{B}	Boltzmann constant, $1.381 \times 10^{-23} \text{ J K}^{-1}$
k_{bg}	Background differential INP concentration.
K	Cumulative nucleus spectrum.
L_{sed}	Loss of insoluble aerosol to sedimentation
M_p	Cold-air outbreak index on pressure level p .
n_{s}	Active site density.
N	Measured aerosol number.
N_0	Empirically derived ice crystal concentration used in early parameterisations of ice number concentration.
N_{aer}	Total measured aerosol number (as a function of size).
$N_{\text{d}}^{\text{obs}}$	Number concentration of cloud droplets. If referring to quantity calculated by the model, this has been adjusted to represent observations.
N_{f}	Number of droplets frozen.
ΔN_{f}	Change in number of droplets frozen over a temperature bin.
$N_{\text{i,con.}}$	Number concentration of ice crystals resulting from contact nucleation.
$N_{\text{i,dep.}}$	Number concentration of ice crystals resulting from deposition nucleation.
$N_{\text{i,s}}^{\text{obs}}$	Number concentration of ice crystals and snow aggregates. If referring to quantity calculated by the model, this has been adjusted to represent observations.
N_{INP}	Number concentration of ice-nucleating particles.
$\bar{N}_{\text{INP}}$	Median number concentration of ice-nucleating particles.
$N_{\text{INP, con.}}$	Number concentration of contact-mode ice-nucleating particles.
$N_{\text{INP, imm.}}$	Number concentration of immersion-mode ice-nucleating particles.
$N_{\text{INP, in hm.}}$	Number concentration of ice-nucleating particles immersed in hydrometeors.
N_{insol}	Number concentration of coarse-mode insoluble aerosol.
$N_{\text{insol, a}}$	Number concentration of coarse-mode insoluble aerosol in air (i.e. not contained in a hydrometeor).
$N_{\text{insol, in hm.}}$	Number concentration of insoluble aerosol in hydrometeors.
$N_{\text{trINP,w}}$	Number concentration of traced INPs in droplets (see Section 5.2).
N_x	Number concentration of modelled hydrometeor species x .
O	Irregularity parameter of a hydrometeor measured by the 2-DS
q_x	Mass mixing ratio of modelled hydrometeor species x .
p_x	Pressure level x .
$\mathbf{r}$	Position vector.
$\bar{r}_{\text{insol}}$	Mean radius of insoluble aerosol.
S	Measured aerosol surface area.

S_{aer}	Total measured aerosol surface area.
S_{i}	Supersaturation with respect to ice.
S_{w}	Supersaturation with respect to liquid water.
t	Time.
Δt	Model timestep.
T	Temperature.
T_0	Triple point of water, $T = 273.15$ K.
T_{act}	Activation temperature of an INP.
T_{cap}	Temperature cap used in the INP-limited INP parametrisation.
T_{cl}	Cloud temperature.
$T_{\mathbf{r}}$	Temperature at grid box with position $\mathbf{r}$.
T_{sub}	$= T_0 - T$.
u	Eastward wind velocity.
$\mathbf{U}$	Horizontal wind vector.
v	Northward wind velocity.
V	Measured aerosol volume.
V_{a}	Volume of air sampled during an aerosol sample.
V_{aer}	Total measured aerosol volume.
V_{d}	Volume of a droplet in a droplet-freezing assay.
x	Placeholder variable for an unspecified CASIM hydrometeor or aerosol tracer.
z	Altitude.
$\mathcal{A}$	Generic aerosol property
$\mathcal{C}_x$	Generic in-cloud property of hydrometeor species x .
$\mathcal{I}$	Generic instrument-measured quantity for comparison with the model.
$\mathcal{M}$	Generic model quantity for comparison with an instrument.
$\mathcal{N}_x$	Particle size distribution of hydrometeor species x .
$\mathcal{P}$	Profile of INP concentration supplied as a tracer in Chapter 5.
$\mathcal{S}$	Number of ice nucleation sites on INPs which have not yet frozen.
$\mathcal{V}_{\text{i}}$	Volume of a water molecule in ice.
α	Fraction of aerosol which is insoluble.
ζ_{i}	Cloud ice fraction.
η_{c}	Efficiency of contact freezing (model parameter).
η_{i}	Efficiency of immersion freezing (model parameter).
θ	Potential temperature
θ_{skt}	Surface skin potential temperature
Θ_{c}	Contact angle between a spherical ice embryo and ice-nucleating surface.
κ	Kinetic ‘pre-factor’ in classical nucleation theory.
λ_x	Slope parameter of particle size distribution for species x .
$\dot{\mu}_{\text{i}}$	Chemical potential associated with a water molecule in the ice phase.

$\dot{\mu}_w$	Chemical potential associated with a water molecule in the liquid phase.
μ_x	Shape parameter of particle size distribution for species x .
ρ	Density of air.
ρ_I^{obs}	Ice water content. If referring to quantity calculated by the model, this has been adjusted to represent observations.
ρ_L^{obs}	Liquid water content. If referring to quantity calculated by the model, this has been adjusted to represent observations.
ρ_T^{obs}	Total water content. If referring to quantity calculated by the model, this has been adjusted to represent observations.
ρ_x	Density (or effective density) of hydrometeor species x .
σ	Standard error.
σ_{insol}	Modal width of coarse mode insoluble aerosol.
σ_{iw}	Surface tension of an ice-water interface.
σ_x	Scale parameter of particle size distribution for species x .
τ	Relaxation constant for nudging of ice crystal concentrations.
ϕ	Latitude.
φ	Efficiency of heterogeneous ice nucleation.
Φ	Relative humidity.
χ	Maximum fraction of insoluble aerosol permitted to freeze (in some CASIM ice nucleation parameterisations).
ψ	Drop-off rate in INP concentration (see Section 4.4.2).

Chapter 1

Introduction

Abstract

Mixed-phase clouds are clouds composed of ice particles and liquid droplets. They are important for Earth's climate system but poorly understood and difficult to model. A significant challenge in representing mixed-phase clouds in models is understanding the role of ice-nucleating particles (INPs), aerosols that permit the heterogeneous freezing of liquid water droplets at temperatures greater than -35°C . Though INPs are key to ice processes in clouds, the global distributions of INP sources, concentrations and sinks are poorly constrained, while INPs and ice nucleation processes are treated in a wide range of ways in models. In this chapter, I introduce mixed-phase cloud processes, particularly considering ice nucleation and the role of INPs in mixed-phase clouds. I describe the way that INPs are modelled and introduce cold-air outbreaks (CAOs), weather systems that create characteristic mixed-phase clouds that form a useful natural laboratory to understand the role of INPs in mixed-phase clouds. I then describe the research questions that this thesis seeks to address by measuring and modelling INPs in CAOs.

1.1 Mixed-phase clouds and the Earth system

Clouds scatter shortwave radiation, and absorb and re-emit longwave radiation (Lohmann, 2016). The net effect of the interactions between clouds and radiation reduces the net radiative flux at the top of Earth's atmosphere by approximately 20 W m^{-2} , cooling the Earth (Wild et al., 2019). This change in flux is called the cloud radiative effect (CRE). In comparison to the CRE, the increase in radiative flux due to anthropogenic greenhouse gas emissions is one-tenth of this magnitude (Forster et al., 2021). Clouds are therefore a crucial component of Earth's energy budget, and it is therefore important to reduce uncertainties in CRE. Clouds may be entirely formed of liquid droplets, entirely formed of ice particles, or a combination of both. This latter group of clouds are termed **mixed-phase** and are found across

the globe in a wide variety of weather systems (Korolev et al., 2017). Mixed-phase clouds account for up to 30 % of global cloud cover (Warren et al., 1988), and data from satellite retrievals suggests that mixed-phase clouds contribute 15–20 % of the global shortwave and longwave CREs (Matus and L'Ecuyer, 2017). The liquid droplets and ice particles in mixed-phase clouds undergo a large set of complex interactions with each other (Morrison et al., 2020). Additionally, mixed-phase clouds undergo many complex interactions with **aerosol**, the suspension of nanometre- to millimetre-sized particles in air (Carslaw, 2022a). This complexity makes it difficult to represent both microphysical and macrophysical properties of mixed-phase clouds such as precipitation rate and albedo in computer models at all scales (Morrison et al., 2020). Given both the difficulty of modelling mixed-phase clouds and their prevalence and importance to Earth's climate, mixed-phase clouds create significant uncertainties not just in numerical weather predictions of clouds in today's climate, but in the modelling of cloud feedbacks — the impact of changes to clouds in a warming climate on the rate of global warming.

1.1.1 Clouds, radiative forcings and climate feedbacks

Radiative forcings describe instantaneous changes in net downward radiative flux at the top of the atmosphere due to a (typically anthropogenic) change to the Earth system. Here, instantaneous means that all other components of the Earth system, such as surface temperature, remain unchanged. Causes of radiative forcings include the emission of greenhouse gases, the emission of aerosol and the change in planetary albedo due to deforestation (Forster et al., 2021; Carslaw, 2022a). Aerosol-cloud interactions, the influence of aerosols on clouds by acting as cloud condensation nuclei (CCN) and as INPs, are an important radiative forcing. Two examples of aerosol-cloud interactions are the Twomey effect and the Albrecht effect. The Twomey effect describes how increased concentrations of CCN results in decreased cloud droplet radii, increasing cloud optical depth and cloud albedo (Twomey, 1974). The consequent reduction in rainfall increases cloud lifetime and the planetary albedo and is referred to as the Albrecht effect (Albrecht, 1989). Since these changes affect the climate system, effective radiative forcing (ERF) is used as a practical way to compare the magnitude of radiative forcings. Calculations of ERF in models permit short-term adjustments to the climate such as the Twomey and Albrecht effects, but hold sea surface temperature constant (Carslaw, 2022a; Sherwood et al., 2020). Overall, aerosol-cloud interactions are the largest source of uncertainty in modelled ERF (Carslaw et al., 2013; Forster et al., 2021), as shown in Figure 1.1.

In contrast, **climate feedbacks** are changes in the Earth system resulting from a change in the planetary temperature which dampen or enhance the global warming that arises from anthropogenic radiative forcing. While the forcings that result in feedbacks are anthropogenic, the feedbacks involve natural processes. Figure 1.2a shows cloud feedbacks grouped by cloud type. As with radiative forcings, climate feedbacks associated with cloud changes are the most uncertain (Forster et al., 2021). In a warming climate, changes to

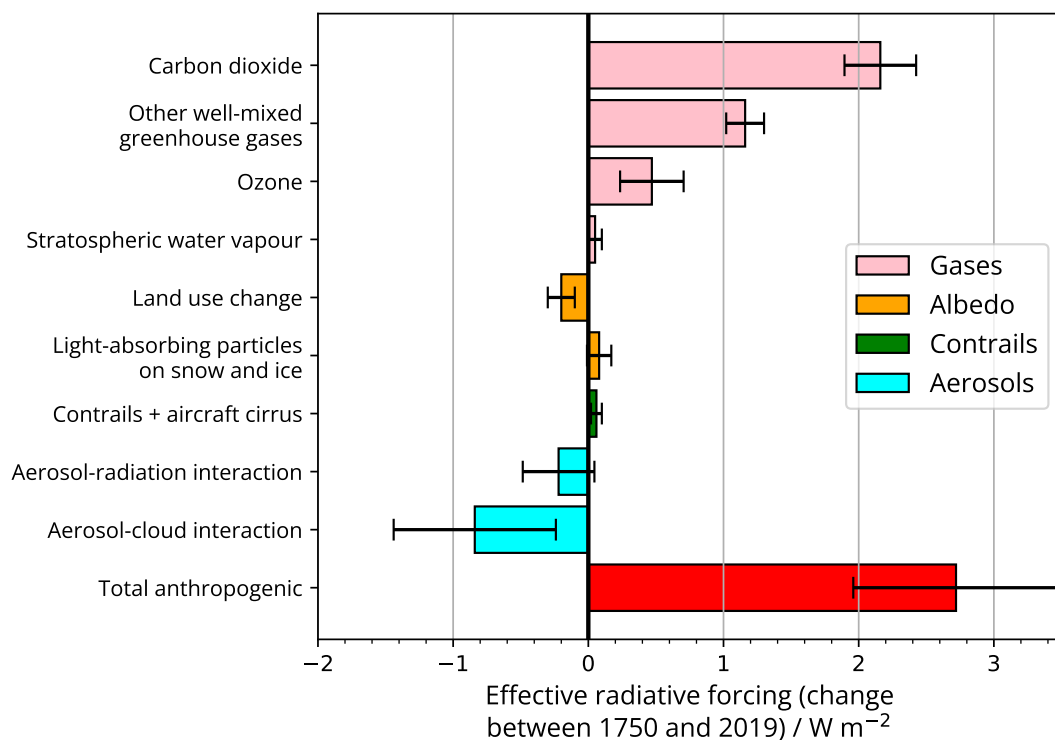


Figure 1.1: The strength of the main anthropogenic radiative forcings as defined by the Sixth Assessment Report of the Intergovernmental Panel on Climate Change (Forster et al., 2021). The error bars depict 5 % to 95 % confidence intervals.

the amount, location, shortwave scattering and longwave emissivity of clouds can change. While the net cloud feedback is positive, it can be decomposed into several components affecting different cloud regimes, as shown in Figure 1.2b. For instance, high clouds are subject to competing feedbacks: a positive feedback associated with an increase in cloud-top altitude without a corresponding increase in cloud-top temperature and subsequent longwave emissivity (Hartmann and Larson, 2002; Forster et al., 2021); and a negative feedback in the tropics associated with the reduction in anvil cloud amount outflowing from deep convective clouds (Bony et al., 2016; Forster et al., 2021).

A highly uncertain feedback pertaining to mixed-phase clouds is the cloud-phase feedback (described further in Figure 1.7). Here, rising isotherms in a warming climate result in the replacement of ice by liquid water. This adjustment results in an increase in the shortwave reflectivity of mixed-phase clouds, which causes a negative climate feedback (Storelvmo et al., 2015). However, the poor representation of mixed-phase clouds in models makes it difficult both to establish the base state of the climate and the magnitude of a future change.

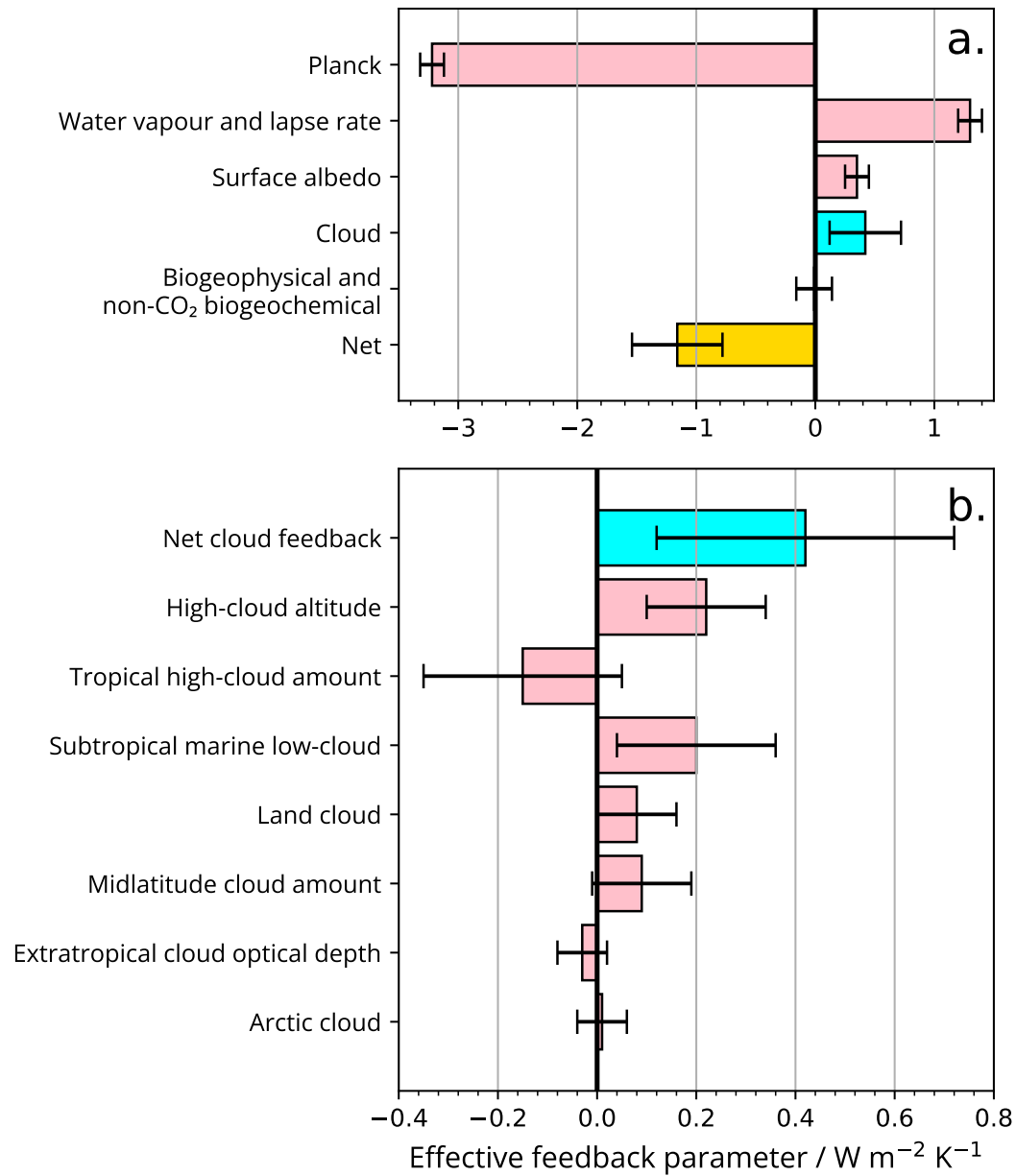


Figure 1.2: The relative strength of climate and cloud feedbacks. Panel (a) shows the strength of the net cloud feedback relative to other major feedbacks and the overall feedback, while panel (b) shows the strength of feedbacks relating to individual cloud regimes (as defined in the Sixth Assessment Report of the Intergovernmental Panel on Climate Change) relative to the total net cloud feedback. Error bars represent uncertainties of one standard deviation from the mean. Adapted from Forster et al. (2021).

1.2 The formation of ice in clouds

In clouds, ice particles may form through primary ice nucleation or secondary ice production. Primary production of ice, or **ice nucleation**, is defined by Vali et al. (2015) as *the first appearance of a thermodynamically stable ice phase*. This contrasts with **secondary ice production** (SIP) where additional ice particles are generated from one or more existing particles. Primary ice nucleation may occur homogeneously, where ice freezes without the presence of an INP, or heterogeneously, where an INP facilitates the freezing. At the temperatures of typical mixed-phase clouds, heterogeneous ice nucleation is the dominant primary ice production mechanism.

1.2.1 Homogeneous ice nucleation

In the absence of an INP, **homogeneous ice nucleation** occurs when molecules in the metastable supercooled liquid phase spontaneously form ice embryos, thermally unstable solid clusters of water molecules (Pruppacher and Klett, 1997; Ickes et al., 2015; Lohmann, 2016). This process can be described using classical nucleation theory, which indicates that for an embryo to form, there is an associated change in the Gibbs free energy, ΔG . This change,

$$\Delta G = \underbrace{\frac{4\pi a_i^3}{\mathcal{V}_i(T)} [\dot{\mu}_i(T) - \dot{\mu}_w(T)]}_{\text{volume term}} + \underbrace{4\pi a_i^2 \sigma_{iw}(T)}_{\text{surface term}}, \quad (1.1)$$

is formed of a volume term and a surface term. The volume term, a negative term representing an energy gain, arises from the difference between the chemical potentials of the ice phase, $\dot{\mu}_i$, and water phase $\dot{\mu}_w$, terms which depend on the temperature, T . This is multiplied by the number of molecules in the embryo, calculated from the radius of the embryo a_i and volume of a water molecule in ice, $\mathcal{V}_i$. By considering the equilibrium between the ice germ and the surrounding droplet, Pruppacher and Klett (1997) showed that this can be expressed as a function of the saturation vapour pressures over water (e_w) and ice (e_i) respectively, such that

$$\dot{\mu}_i(T) - \dot{\mu}_w(T) = -k_B T \ln \left(\frac{e_w(T)}{e_i(T)} \right). \quad (1.2)$$

The surface term, a positive term representing an energy cost, describes the energy required to form the water-ice interface of the embryo. This is calculated by multiplying the spherical surface area of the embryo by the surface tension between ice and water, σ_{iw} . The competition between these terms creates a critical radius a_{crit} of nucleation at which ΔG is maximal. Finding a_{crit} by differentiating ΔG with respect to a_i allows us to calculate the energy barrier to germ formation, ΔG^* ,

$$\Delta G(a_{\text{crit}}) \equiv \Delta G^* = \frac{16\pi \mathcal{V}_i \sigma_{iw}}{3 [k_B T \ln(e_w/e_i)]^2}. \quad (1.3)$$

Using the Boltzmann distribution, the number of germs can be calculated using ΔG^* . Multiplying this by a kinetic pre-factor, κ , which incorporates the diffusive flux of water molecules available for the ice germ and the activation energy for the transfer of a water molecule across the water-ice boundary, leads to a steady-state nucleation rate for homogeneous nucleation, J_{hom} ,

$$J_{\text{hom}} = \kappa \exp\left(\frac{\Delta G^*}{k_B T}\right). \quad (1.4)$$

A significant rate of homogeneous ice nucleation is not met until temperatures are below approximately -35°C to -38°C (Pruppacher and Klett, 1997; Ickes et al., 2015; Koop and Murray, 2016). As such, homogeneous nucleation is typically an largely unimportant process in mixed-phase clouds — at temperatures low enough for appreciable homogeneous nucleation to occur, homogeneous nucleation is too rapid for liquid water to be sustained. However, the parameters of J_{hom} are subject to uncertainty (Ickes et al., 2015) and using estimates at the limits of the uncertainty range which give the highest feasible homogeneous nucleation rate, homogeneous freezing could occur at -30°C , within the typical mixed-phase cloud temperature range (Herbert et al., 2015).

Theoretically, it is also possible for ice to nucleate homogeneously directly from water vapour, a process known as **homogeneous deposition nucleation** (not to be confused with *deposition nucleation*, which refers to the heterogeneous process described in Section 1.2.2) (Murray and Jensen, 2010). However, this would require extremely high supersaturations that are not observed in the atmosphere (Pruppacher and Klett, 1997; Lohmann, 2016). In the upper layers of the atmosphere, such as the mesosphere, it has been theorised that homogeneous deposition nucleation could occur via an intermediate stage of liquid water formation (Murray and Jensen, 2010). However, other theoretical studies suggest that any such nucleation would be suppressed by heterogeneous nucleation even if possible (Tanaka et al., 2022) and this process has never been observed (Vali et al., 2015).

1.2.2 Heterogeneous ice nucleation

At temperatures higher than -35°C , the presence of newly formed ice requires heterogeneous freezing, where an INP provides a favourable site for nucleation, reducing the energy barrier for nucleation to occur. There are several modes of **heterogeneous ice nucleation**, which are depicted in Figure 1.3. Historically, there has been debate over the definitions of these modes, particularly where they are difficult to differentiate observationally (Kanji et al., 2017). To describe them here, I refer to definitions presented by Vali et al. (2015) that were agreed through community consensus in an open, interactive community peer-review process (Carslaw, 2015).

Deposition nucleation occurs when ice nucleates directly onto an INP from water vapour when water vapour is supersaturated with respect to ice. By definition, there is no prior formation of liquid in this mode of nucleation (Vali et al., 2015), though observations by

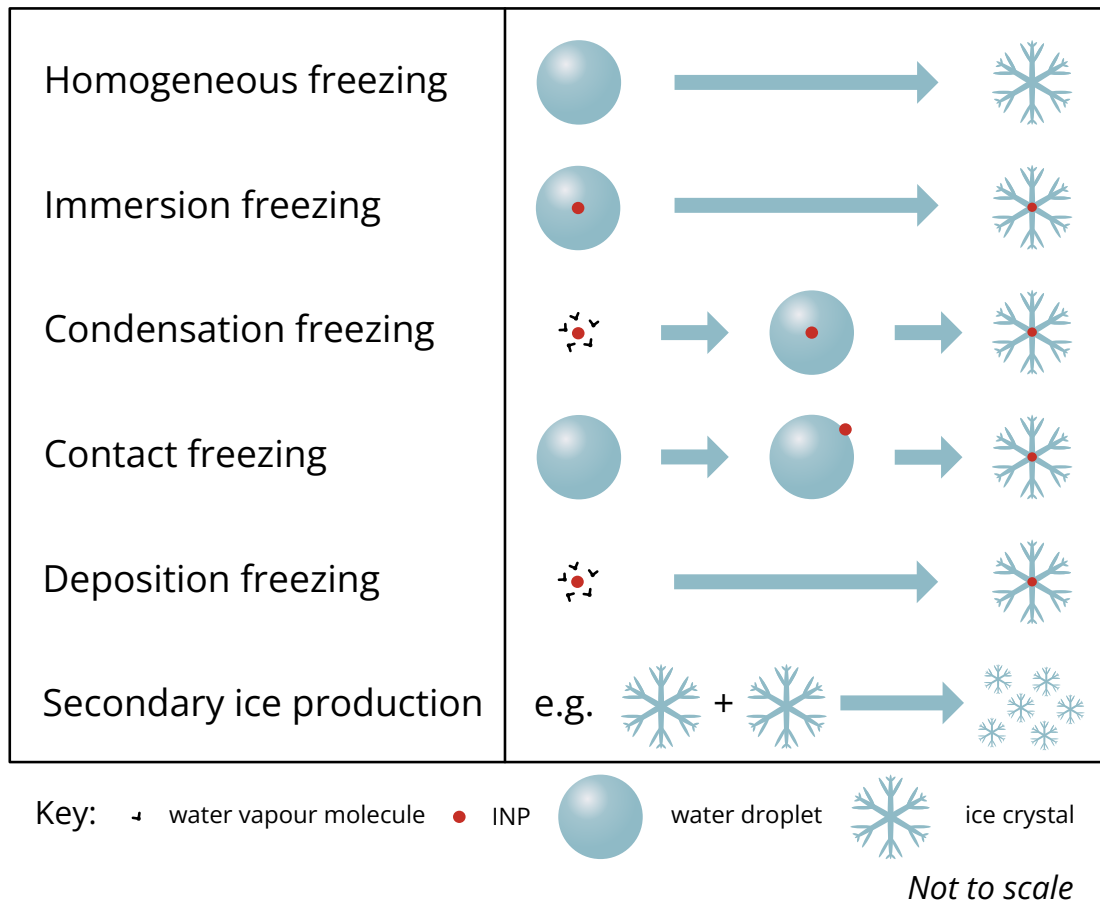


Figure 1.3: Mechanisms by which ice can form in clouds, adapted from a schematic by Kanji et al. (2017). Secondary ice production is depicted using the example of ice-ice collisions, but this is one of many mechanisms.

Marcolli (2014) indicate that some nucleation that is commonly identified as deposition nucleation may occur when water in cavities on the surface of an INP freezes. **Immersion freezing** occurs when ice nucleation is initiated by an INP that is contained within a liquid droplet (Vali et al., 2015). This is a similar process to **condensation freezing**, which is defined by Vali et al. (2015) as freezing that is *initiated concurrently with the initial formation of liquid on an INP that acts as a cloud condensation nucleus*. Despite the differing definitions, there is some debate as to whether or not immersion freezing and condensation freezing should be considered to be the same mode of nucleation (Kanji et al., 2017). Immersion freezing is the most important heterogeneous freezing mode in mixed-phase clouds, which often have supercooled liquid layers and ice-free regions which are supersaturated with respect to ice (Murray et al., 2012b). The final mode of nucleation, **contact freezing**, occurs when an INP makes contact with liquid water at the air-liquid interface of a liquid droplet. This can occur from the inside-out if a droplet begins to evaporate allowing the surface of a drop to meet the interface (Durant and Shaw, 2005). Although heterogeneous nucleation is intuitively associated with mixed-phase clouds (where homogeneous nucleation does not typically occur), heterogeneous nucleation also occurs at temperatures below -38°C for some types

of INP (Section 1.2.3) and can outcompete homogeneous nucleation at these low temperatures in some circumstances (Cziczo et al., 2013). In this work, I will focus on heterogeneous freezing in the mixed-phase cloud regime.

1.2.2.1 The stochastic and singular hypotheses

The processes of heterogeneous ice nucleation can be described by either a stochastic, time-dependent model or a singular time-independent model, with evidence supporting both approaches (Vali, 2014). The **stochastic** hypothesis assumes that within an INP population, the sites are of equal effectiveness and that freezing occurs at a rate that is dependent on the number of sites and the temperature (Westbrook and Illingworth, 2013; Vali et al., 2015). This can be represented by an extension to classical nucleation theory, calculating ΔG_{het}^* , the nucleation energy barrier of heterogeneous nucleation, as

$$\Delta G_{\text{het}}^* = \varphi \Delta G^*, \quad (1.5)$$

where φ is the ice nucleation efficiency, a value between 0 and 1, where 0 represents a perfect ice-nucleator¹ and 1 represents a surface with no ice-nucleating ability (Murray et al., 2012b). For modes other than deposition nucleation, φ can be expressed as a function of the contact angle, Θ_c between a spherical ice embryo and the surface,

$$\varphi = \frac{(2 + \cos \Theta_c)(1 - \cos \Theta_c)^2}{4}. \quad (1.6)$$

Since the critical ice cluster may not be spherical, Murray et al. (2012b) recommend that Θ_c be regarded as a *semi-empirical measure of how well a material catalyses ice nucleation*.

However, it is known that not all sites of ice nucleation on an INP are of equal ice-nucleating ability, and that ice will preferentially nucleate at sites with higher activation temperatures (Holden et al., 2019). The **singular** hypothesis of ice nucleation represents this heterogeneity using the cumulative nucleus spectrum, $K(T)$, an empirically derived function of temperature describing the number concentration of sites (Vali et al., 2015). Experimentally, this is typically derived from a droplet-freezing assay, where droplets of pure water containing aerosol particles from a sample are cooled and the temperature at which droplets freeze are recorded (see Section 2.2.2 for a description of techniques and Section 2.3.1 for a comparison in the context of Arctic INP measurements). To do so, the method of Vali (1971) is used, where the differential nucleus spectrum $k(T)$ is calculated as

$$k(T) = \frac{-1}{V_d (1 - f_f(T))} \frac{df_f}{dT}, \quad (1.7)$$

where V_d is the volume of a droplet and f_f is the fraction of all droplets that frozen. This is

¹ such as ice-nine (Vonnegut, 1963).

integrated with respect to temperature to obtain the cumulative nucleus spectrum,

$$K(T) = \int_0^{f_t} k(T) dT = -\frac{1}{V_d} \ln [1 - f_f(T)]. \quad (1.8)$$

To compare the effectiveness of aerosol populations as ice nucleators, this is typically normalised by the surface area of sampled aerosol (A_{sample}) to obtain n_S , the active site density,

$$n_S = \frac{K(T)}{A_{\text{sample}}}. \quad (1.9)$$

According to the singular hypothesis, the freezing is deterministic and therefore independent of time (Westbrook and Illingworth, 2013; Vali, 2014; Vali et al., 2015). Like the stochastic hypothesis, this fails to fully represent observations of freezing, but in many cases, the temperature dependence of sites dominates the time-dependence of nucleation since the cooling rate in clouds is less than the rate of nucleation (Vali, 2008; Herbert et al., 2014; Holden et al., 2019).

As well as the singular and stochastic hypotheses, there are also descriptions that attempt to reconcile the two approaches, such as the “soccer-ball” model of nucleation proposed by Niedermeier et al. (2011) or the Framework for Reconciling Observable Stochastic Time-dependence of Herbert et al. (2014).

1.2.3 Types of ice-nucleating particles

There are many types of aerosol which are able to nucleate ice. In mixed-phase clouds globally, **mineral dust** is thought to be one of the most important INP types owing to its abundance and high ice-nucleating ability (Murray et al., 2012b; Boose et al., 2016). Emissions of dust happen from almost all land regions around the world, but the greatest emission sources are large arid deserts such as the Sahara and Gobi (Kok et al., 2021a, 2023). Dust from these strongest source regions can be transported across continents and oceans and are able to act as INPs far from their source regions (Creamean et al., 2013, 2014; Kok et al., 2021b). Additionally, there are significant emissions of dust outside of large desert regions, including in the high latitudes (Bullard et al., 2016). Different dust minerals have different ice-nucleating abilities, with potassium feldspar thought to be the most effective ice-nucleating dust mineral, causing appreciable amounts of ice nucleation at temperatures below -15°C (Atkinson et al., 2013; Harrison et al., 2019).

Many **bioaerosols** are also effective INPs. Over 300 different biological species have been identified as ice-nucleators, including bacteria, fungi, pollens, mosses, planktons and lichens (Huang et al., 2021; Tarn et al., 2025). Some bioaerosols, particularly bacteria such as *Pseudomonas syringae*, are able to nucleate ice at extremely high temperatures ($-3^\circ\text{C} \leq T \leq 0^\circ\text{C}$) (e.g. Maki et al., 1974; Vali et al., 1976). While bioaerosols may nucleate ice, biological components of aerosols can also enhance the ability of mineral dusts to nucleate ice (Herbert

et al., 2025). Agricultural (or **fertile**) **soil dusts**, which can carry a biogenic component, have been shown to have higher active site densities than dust from arid regions (Steinke et al., 2016). This enhancement of ice-nucleating ability is attributed to the presence of nano-scale fragments of biological material, such as protein macromolecules, which can be adsorbed to the surface of mineral dusts (Pummer et al., 2012; O’Sullivan et al., 2015; O’Sullivan et al., 2016). Similarly, biogenic material from **sea-spray** is able to nucleate ice, though its ice-nucleating ability is typically lower than that of mineral dusts (Wilson et al., 2015; DeMott et al., 2016; McCluskey et al., 2018b). As such, sea spray INPs are typically most important in remote marine regions where the dust concentration is particularly low (Vergara-Temprado et al., 2017; Huang et al., 2018; Herbert et al., 2025).

There are many other types of INPs, but these are typically less important for mixed-phase clouds. For instance, **black carbon** is an effective ice-nucleator in the cirrus regime (Kanji et al., 2017), but at warmer temperatures it is of second-order importance at most, despite its abundance (Vergara-Temprado et al., 2018a; Kanji et al., 2020). Other INP types include metal oxides (typically from anthropogenic activities), glassy organic aerosols and ash from volcanic eruptions (Kanji et al., 2017).

1.2.4 Secondary ice production

Though the processes of heterogeneous ice nucleation are important, atmospheric ice particle concentrations may be orders of magnitude larger than INP concentrations. This discrepancy is often attributed to mechanisms of secondary ice production in which new ice particles are created from existing ice particles without the involvement of a further INP. A wide variety of SIP mechanisms have been observed and studied in the laboratory but their relative importance for ice particle number concentration is unknown and the mechanisms are difficult to represent in models (Field et al., 2017; Korolev and Leisner, 2020). Several of these mechanisms are depicted in Figure 1.4.

The best-studied SIP mechanism is the Hallett-Mossop (HM) process, where small splinters of ice are ejected from ice particles undergoing the process of riming (growth by sweeping up supercooled liquid droplets, Section 1.3.2) (Hallett and Mossop, 1974). The HM process occurs predominantly between the temperatures of -3°C and -8°C (Field et al., 2017). Although the HM process is one of the best-documented secondary ice processes, a recent laboratory experiment by Seidel et al. (2024) failed to observe significant ice multiplication through the HM process. However, this experiment has been criticised for its representation of graupel particles and using droplets too large (Biggart et al., 2026).

Secondary ice production also occurs by a wide range of other mechanisms (Field et al., 2017; Korolev and Leisner, 2020). Splinters of secondary ice may be produced from collisions of ice particles with other ice particles (Vardiman, 1978) or by collisions of supercooled liquid droplets with ice particles (James et al., 2021). Ice splinters are also formed when a droplet shatters during the process of immersion freezing if the initial droplet is sufficiently

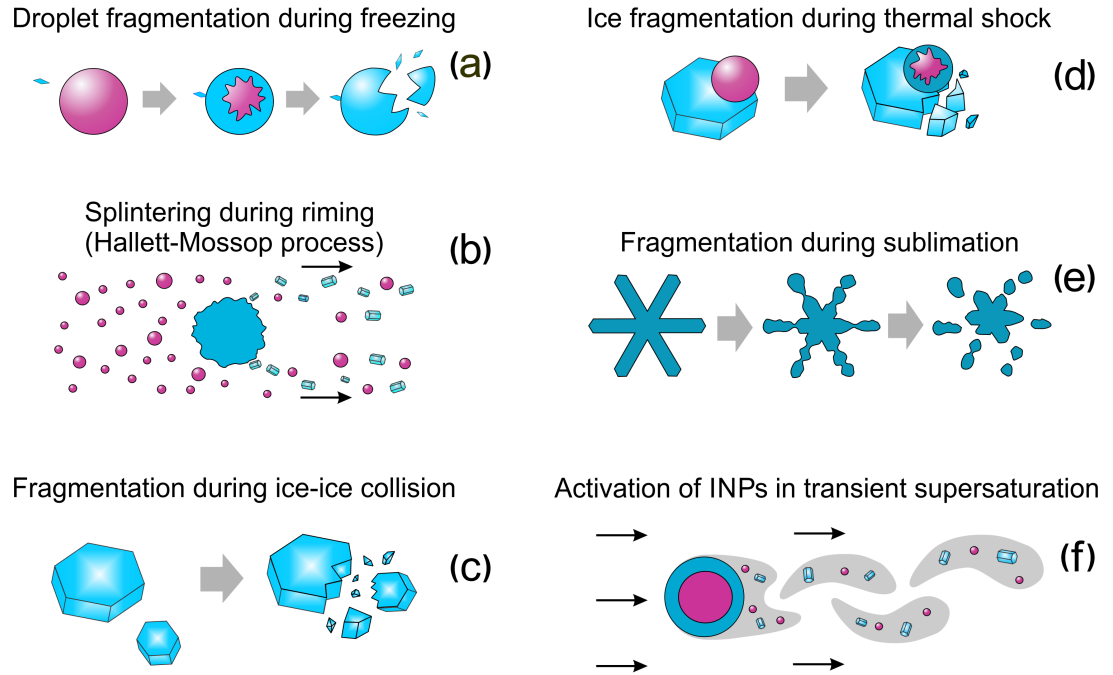


Figure 1.4: A schematic of several SIP mechanisms. Reproduced with permission from Korolev and Leisner (2020) under a CC-BY-4.0 license.

large (Lauber et al., 2018). Similarly, thermal shocks associated with the latent heat transfer between a drop riming on an ice particle may cause the droplet to shatter (Korolev and Leisner, 2020). Alternatively, ice particles may fracture into smaller parts during the process of sublimation (Oraltay and Hallett, 1989), though this is unlikely to be important for clouds (Korolev and Leisner, 2020).

1.2.5 Representing ice formation in models

It is clearly far too computationally expensive to track individual aerosol particles explicitly in atmospheric models, so freezing must be parameterised based on laboratory measurements and observations from field studies. Over the last sixty years, the complexity of freezing parameterisations has increased and such parameterisations have become increasingly effective, though still imperfect (Herbert et al., 2025).

The first freezing parameterisations were very simple and related ice particle concentration directly to temperature and INP concentration without making explicit connection to the nature of the ice-nucleating particles themselves. Fletcher's 1962 parameterisation only considered deposition-mode nucleation, with an exponential relationship

$$N_{i,\text{dep.}} = N_0 \exp(aT_{\text{sub}}), \quad (1.10)$$

between the ice particle concentration $N_{i,\text{dep}}$ and the temperature below the triple point of water, T_{sub} , where N_0 is an empirically derived ice nucleus concentration and a is a fitting

parameter based on ice particle measurements in the environment.

However, this significantly underestimated ice concentrations at temperatures greater than -25°C (Meyers et al., 1992). A decade later, Young (1974) developed a theoretical model for contact nucleation based on Brownian motion, and parameterised it such that

$$N_{\text{i,con.}} = N_0(z) (b - T)^a, \quad (1.11)$$

where b is a fitting parameter and N_0 depends on altitude, z , and ranged from 200 L^{-1} at sea level to 10 L^{-1} at 5000 m above sea level. The first parameterisation of both deposition and contact nucleation was created by Cotton et al. (1986) who added a supersaturation dependence into Fletcher's deposition scheme, such that

$$N_{\text{i,dep.}} = N_0 \left[\frac{S_{\text{i}} - 1}{S_{\text{i}}/S_{\text{w}} - 1} \right]^b \exp(aT_{\text{sub}}), \quad (1.12)$$

where S_{w} and S_{i} are supersaturations with respect to liquid water and ice. Meyers et al. (1992) made further developments on these parameterisations. They adjusted the Cotton parameterisation so that deposition nucleation depended only on ice supersaturation,

$$N_{\text{i,dep.}} = N_0 \exp[a + b(S_{\text{i}} - 1)], \quad T < -5^\circ\text{C} \quad (1.13)$$

providing good matches to observed ice particle concentrations at temperatures between -7°C and -20°C . Meyers et al. also adjusted the Young contact nucleation scheme to reflect a smaller average INP size than previously thought, providing an upper limit on contact nucleation at

$$N_{\text{i,con.}} = \exp(a + bT_{\text{sub}}), \quad T < -2^\circ\text{C}, \quad (1.14)$$

with the exact ice particle concentration in a model moderated by collection rates of droplets.

More recent approaches account for the nature of ice-nucleating particles rather than a number of ice nuclei, either with a singular or stochastic approach. Singular parameterisations of freezing using a surface density include those of Connolly et al. (2009), Niemand et al. (2012) and Atkinson et al. (2013) for mineral dusts analysed in the laboratory. These take the general form

$$N_{\text{INP}} = f(T, S_{\text{i}})g(\mathcal{A}), \quad (1.15)$$

where f is a function of temperature (and sometimes supersaturation) and g is a function of $\mathcal{A}$, a property of the aerosol types considered, such as surface area or number concentration. Singular parameterisations which take into account more than one type of aerosol can be derived from observational measurements. A particularly widely used singular INP parameterisation based on observations is the DeMott et al. (2010) parameterisation, which predicts an INP concentration based on the number of aerosol with diameters larger than 500 nm. Such parameterisations can be applied to modelling cases where the sources

and nature of INPs are better-understood, with the INP parameterisation chosen to reflect observations relevant to the case (Gjelsvik et al., 2025). These parameterisations are particularly advantageous in modern aerosol-aware models as they are able to diagnose INP concentrations from aerosol tracers in a model, thus allowing for spatial variability in INP concentration at a given temperature, rather than this being fixed across the model domain as in the Meyers and Cotton parameterisations. Many similar parameterisations have been developed for more specific cases, such as DeMott et al. (2015), which considers desert dust and McCluskey et al. (2018b), which considers marine organic aerosols from sea-spray. Alternatively, stochastic parameterisations include that of Hoose et al. (2010), while Phillips et al. (2008) applies this approach to a variety of aerosol types. parameterisations with multiple contact angles can be used to describe INPs with heterogeneous surfaces (Kanji et al., 2017).

There are also approaches which combine the stochastic and singular approaches. Vali (1994) proposed a modified singular approach, adding an offset to the temperature in Eq. 1.15 based on the cooling rate, an approach used in the parameterising of the ice-nucleating ability of kaolinite by Murray et al. (2011). The “soccer-ball” reconciliation of singular and stochastic treatments (Niedermeier et al., 2011) has also been simplified for use in models (Niedermeier et al., 2014). In this model, each particle is divided into a number of surface sites with different J_{het} functions that are assigned a random fixed contact angle according to a Gaussian distribution.

While many parameterisations have been developed to study primary ice production, with varying degrees of success, SIP is rarely parameterised in models beyond formulations for the HM process (Field et al., 2017). Typically, parameterisation of other methods of SIP is constrained to process studies such as that of Sotiropoulou et al. (2021), who studied mechanical breakup of ice particles.

1.3 The behaviour of ice in clouds and the influence of INPs

Once ice has nucleated in a cloud, it will interact with water vapour and existing hydrometeors in the cloud, as depicted in Figure 1.5.

1.3.1 Diffusional growth and the Wegener-Bergeron-Findeisen process

When ice exists in a mixed-phase cloud, and the vapour pressure is supersaturated with respect to ice, ice crystals will grow through the deposition of water vapour molecules onto the crystal. This process is known as **diffusional growth**. When the vapour pressure is also subsaturated with respect to water (which is the case when the vapour pressure is slightly supersaturated with respect to ice), liquid droplets in a mixed-phase cloud will evaporate, depleting the mass of liquid water. The rate of this process is driven by the difference in saturation vapour pressure over ice and water (Wegener, 1911). It is possible for ice crystals to

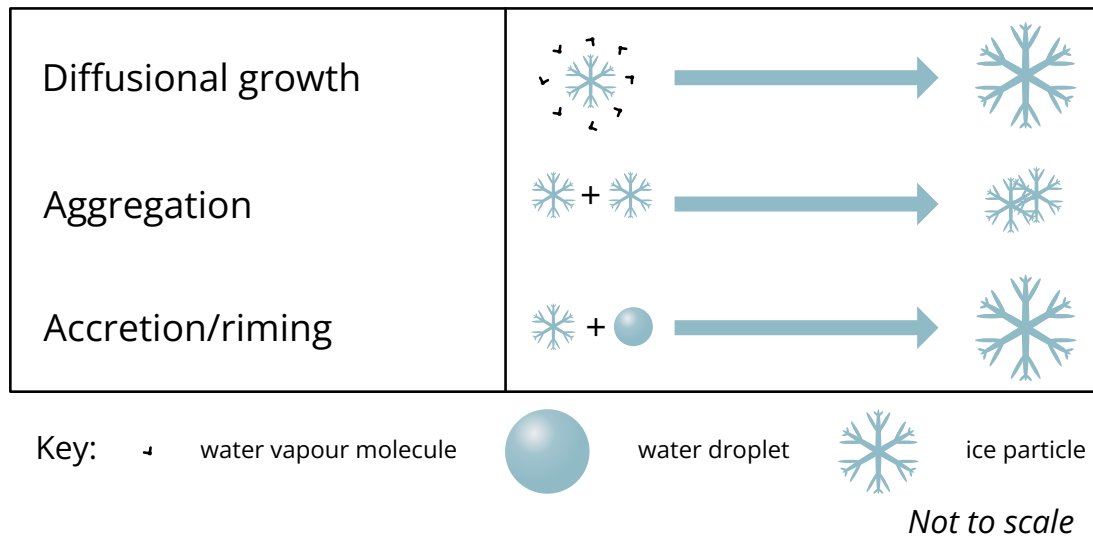


Figure 1.5: Mechanisms by which ice can grow once nucleated in a cloud. Based on descriptions in Rogers and Yau (1989), Pruppacher and Klett (1997), Lohmann (2016) and Khain and Pinsky (2018).

grow through this process to sizes at which they can precipitate (Bergeron, 1935; Findeisen, 1938). This mechanism is known as the Wegener-Bergeron-Findeisen (WBF) process. As such, an increased ice concentration resulting from an increased INP concentration in a mixed-phase cloud can result in an enhancement of precipitation.

1.3.2 Collisional growth of ice

As well as growing from water vapour, ice can grow by colliding with other hydrometeors in the cloud, in processes akin to collision-coalescence between liquid droplets (Khain and Pinsky, 2018). As for liquid droplets, collisional growth of ice particles is driven by the difference in fall speeds between two hydrometeors (Pruppacher and Klett, 1997), and can also occur when particles collide due to turbulence (Lohmann, 2016). **Aggregation** occurs when two ice particles collide and stick together and is responsible for the formation of snowflakes (Pruppacher and Klett, 1997). Though aggregation is feasible at temperatures as low as -40°C (Hobbs, 1965), aggregation occurs largely at temperatures higher than -10°C due to the increased thickness of the quasi-liquid layer on the surface of ice particles at these warmer temperatures (Dobrowolski, 1903; Pruppacher and Klett, 1997; Lohmann, 2016). **Riming** (also known by the more general term, **accretion** (Lohmann, 2016)) occurs when ice particles collide with supercooled droplets typically of smaller diameter (Rogers and Yau, 1989) and is the process that produces graupel and hail (Reinking, 1975). Relative to the ground, graupel particles may be falling relative while the smaller cloud droplets are moving upwards. However, the slower fall speed of ice particles relative to droplets of the same size gives the opportunity for ice particles to grow much larger than droplets through an increased number of collisions (Lohmann, 2016).

In atmospheric models, **self-collection** (particles of similar nature colliding and aggregating)

is often distinguished from aggregation of particles with different sizes (Khain and Pinsky, 2018). Where multiple species of ice with different sizes are traced in a model (for instance ice crystals and snowflakes) the parameterised process of **autoconversion** is introduced. Autoconversion estimates the change in quantities of both traced species that occur due to collisions of the smaller species that produce particles of the larger species (Khain and Pinsky, 2018).

1.3.3 Ice precipitation

The majority of global precipitation involves clouds containing ice, with Mülmenstädt et al. (2015) finding that even liquid rain normally arises from mixed- or ice-phase cloud. Over extratropical land, the proportion of precipitation events which originate from a cloud containing ice is as high as 99 % (Mülmenstädt et al., 2015). When an ice particle reaches a large enough size via diffusional growth or collisions, the fall speed of the ice particle will exceed the speed of the competing updraught and the ice particle will be removed from the cloud. Most precipitating ice is snow or graupel, with snow dominant in stratiform clouds and the larger, denser graupel particles dominant in convective clouds where updraughts are sufficiently high to inhibit snowfall (Lohmann, 2016). While the majority of precipitating ice forms through collision processes, it is possible (but uncommon) for ice crystals to grow solely through diffusional growth to a size large enough to precipitate even in a largely cloudless sky (Intrieri and Shupe, 2004; Lohmann, 2016). Since ice in mixed-phase clouds is originally initiated through heterogeneous ice nucleation, the precipitation can act as an efficient sink of INP concentrations (Stopelli et al., 2015). Since precipitating ice readily undergoes many collisions with droplets in a mixed-phase cloud, ice precipitation can also act as a strong sink of aerosols which act as CCN rather than INPs. If the precipitation falls into air which is subsaturated with respect to ice, it is possible for snow and graupel to sublimate, returning aerosol to the air. Similarly, snow or graupel falling into air which is warmer than 0 °C will begin to melt. The return of aerosol could then occur through evaporation, or alternatively aerosol (including INPs) could be emitted upon the impact of the raindrop with the ground (Mignani et al., 2025).

1.4 INPs, ice and the macrophysics of mixed-phase clouds

Mixed-phase clouds are defined as those that contain both supercooled liquid droplets and ice particles, and so can be sustained at temperatures between 0 °C and –38 °C. These temperatures are low enough for ice to remain frozen but warm enough that supercooled droplets will not necessarily freeze homogeneously. However, this definition is ambiguous, as depicted in Figure 1.6. Schematic (a) shows the most straightforward conceptual mixed-phase cloud – one where ice and liquid occur in relatively equal amounts and the hydrometeors are approximately homogeneously mixed. In contrast, schematic (b) shows a cloud with a very small amount of liquid and a very large amount of ice. Since both phases are present,

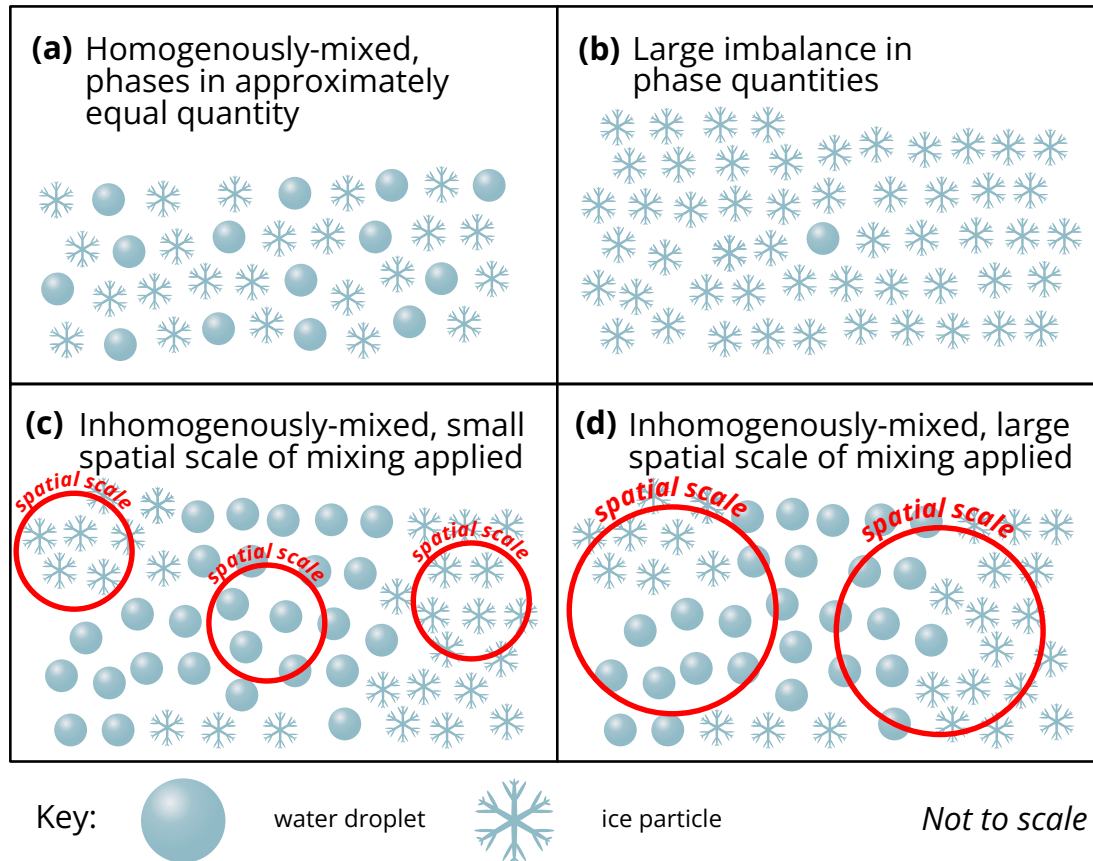


Figure 1.6: Schematics that depict the challenge of defining “mixed-phase”, based on conceptual diagrams in Korolev et al. (2017).

this must be a mixed-phase cloud according to the definition. However, if the quantity of liquid is small enough, it will have no meaningful effect on the cloud, so it could be argued that the cloud should be considered to be in the ice phase.

When clouds are heterogeneously mixed, the spatial scale of mixing relative to the hydrometeor distribution becomes important. This is shown by schematics (c) and (d) of Figure 1.6, which depict the same cloud but apply two different scale lengths of mixing. In case (c), the small spatial scale of mixing means the cloud can be considered to be separate liquid and ice clouds. Conversely, in case (d), the larger scale length of mixing means the cloud can be considered mixed-phase. While cloud parameterisations in global models represent mixed-phase clouds as a homogeneous mix of ice and liquid, Sokol and Storelvmo (2024) showed that there is significant spatial heterogeneity in cloud mixing and hypothesised that this may be related to ice-nucleating particles. Constraining global model parameters to those that better represent the heterogeneity of mixing has been shown to increase projections of climate warming (Hofer et al., 2024). In models on smaller scales, mixing of hydrometeor phases is represented using the mixed-phase overlap fraction (MPOF), which governs the strength of interactions between liquid and ice (Evans et al., 2025).

Unlike for ice nucleation, the community has not agreed on one definition of a mixed-phase

cloud and definitions vary between observational and theoretical perspectives (Korolev et al., 2017). A generic calculation to determine the phase of a cloud based on the relative quantities of ice and water was introduced for in-situ measurements of clouds by Korolev et al. (1998). They calculated ζ_i , the relative fraction of ice, as

$$\zeta_i = \frac{C_i}{C_i + C_w}, \quad (1.16)$$

where C_i is a generic hydrometeor property for ice such as number concentration or mass mixing ratio and C_w is its equivalent for liquid. If ζ_i is between an upper and lower threshold (typically 0.1–0.9 (Korolev et al., 2017)), the cloud is considered mixed-phase. However, this definition does not define a spatial scale of averaging for this calculation. In their review, Korolev et al. (2017) suggest that this is chosen by considering the characteristic timescale of the dominant interaction between ice and liquid in the cloud. For instance, if this is turbulent mixing, then the length scale associated with this should be chosen.

The relative concentrations of liquid and ice control the radiative properties of a mixed-phase cloud, since for a given mass of cloud water, incoming solar radiation is scattered more efficiently when the water is in the ice phase rather than the liquid phase. As such, a mixed-phase cloud with a greater proportion of ice will tend to reflect less incoming short-wave radiation than an equivalent cloud with more liquid (Sun and Shine, 1994, 1995; Garrett et al., 2001), so an increased INP concentration is generally associated with a decrease in mixed-phase cloud reflectivity (Vergara-Temprado et al., 2018b; Murray et al., 2021). However, this is not always the case: investigations of shallow mixed-phase cumulus clouds by Huang et al. (2025) showed that when mixed-phase clouds were initially ice-dominated, increasing the number of INPs resulted in a “Twomey-like” effect for ice. Here, increased ice nucleation reduced the average size of ice particles while increasing their number concentration, thereby increasing the reflectivity of the cloud.

1.4.1 Modelling the effect of INPs on cloud macrophysics

The concentration of INPs has been shown to strongly affect bulk cloud properties in regional modelling experiments. For instance, Hawker et al. (2021) showed that the outgoing radiation from a deep convective cloud was strongly controlled by the strength of the temperature dependence in the INP parameterisation chosen to simulate it. Similarly, Lüttmer et al. (2025) showed that simulations of the structure of a deep convective cloud were dependent on the INP parameterisation chosen. However, increased complexity of INP schemes does not necessarily improve the representation of clouds. In regional modelling of Arctic stratocumulus clouds, Possner et al. (2024) found the differences between clouds modelled using prognostic and diagnostic INP parameterisations were masked by the influence of SIP.

Cloud macrophysics in global models is also sensitive to INPs. For instance, improving the

representation of INPs by relating INPs to observed dust concentrations resulted in an increased cloud radiative effect of 0.05 W m^{-2} to 0.26 W m^{-2} through the depletion of liquid water caused by increased freezing (Shi and Liu, 2019). This figure is comparable to the magnitude of the total aerosol-radiation interaction effective radiative forcing (Figure 1.1), showing the potential for INPs to affect the global climate. However, Shi and Liu (2019) noted that their estimate could be reduced in magnitude or even reversed in direction by increased dust concentrations in the Arctic, and did not include INPs from other sources such as sea-spray. Similarly, using a region-specific INP parameterisation of INPs has been shown to improve the prediction of CRE by models (Gjelsvik et al., 2025). Alongside forcings, estimates of global cloud feedbacks are also affected by the choice of INP parameterisation, which significantly changes the representation of the base state of the climate (McGraw et al., 2023). While it is clearly important to represent INPs correctly to model cloud macrophysics at a global scale, improvements in INP representation still require good representations of other processes. For instance, Sotiropoulou et al. (2024) found that a combination of improved primary and secondary ice production representations was required to improve CRE biases in the NorESM2 climate model, while Imura and Suzuki (2025) found that perturbations of INP concentrations had opposite impacts on CRE when used with two different precipitation schemes in the MIROC6 climate model.

1.4.2 INPs and the cloud-phase feedback

Several modelling studies (e.g. Prenni et al., 2007; Bangert et al., 2012; Vergara-Temprado et al., 2018b; McGraw et al., 2023; Huang et al., 2025) have shown that the reflectivity of a mixed-phase cloud is partly controlled by its INP concentration, as this modulates the concentration of ice. Even in clouds where SIP dominates the ice concentration, SIP is linked to the pre-existing primary ice (Murray et al., 2025). The control of cloud reflectivity by INPs consequently suggests that INPs could modulate the cloud-phase feedback, as shown in Figure 1.7. If two mixed-phase clouds form in identical locations in identical meteorological conditions, it would be expected that the cloud with more INPs has a greater ice particle number concentration and thus a reduced reflectivity. According to the cloud-phase feedback (see Section 1.1), in a warming climate, ice in mixed-phase clouds will be replaced by water, increasing cloud reflectivity. In this case, the cloud with more INPs has a greater potential for the replacement of ice with water, and thus a greater potential negative cloud feedback (Murray et al., 2021).

The type of INPs found in mixed-phase clouds may also affect the strength of this cloud feedback. If the dominant INP type in the cloud has a strong temperature dependence (i.e. a small change in temperature corresponds to a large change in INP concentration), then the future change in cloud reflectivity will also strongly depend on INP concentration as a small temperature increase can result in the replacement of a large amount of ice (Murray et al., 2021). Similarly, future changes in INP emissions may affect the strength of this cloud feedback. For instance, increasing desertification in a warming climate will result in

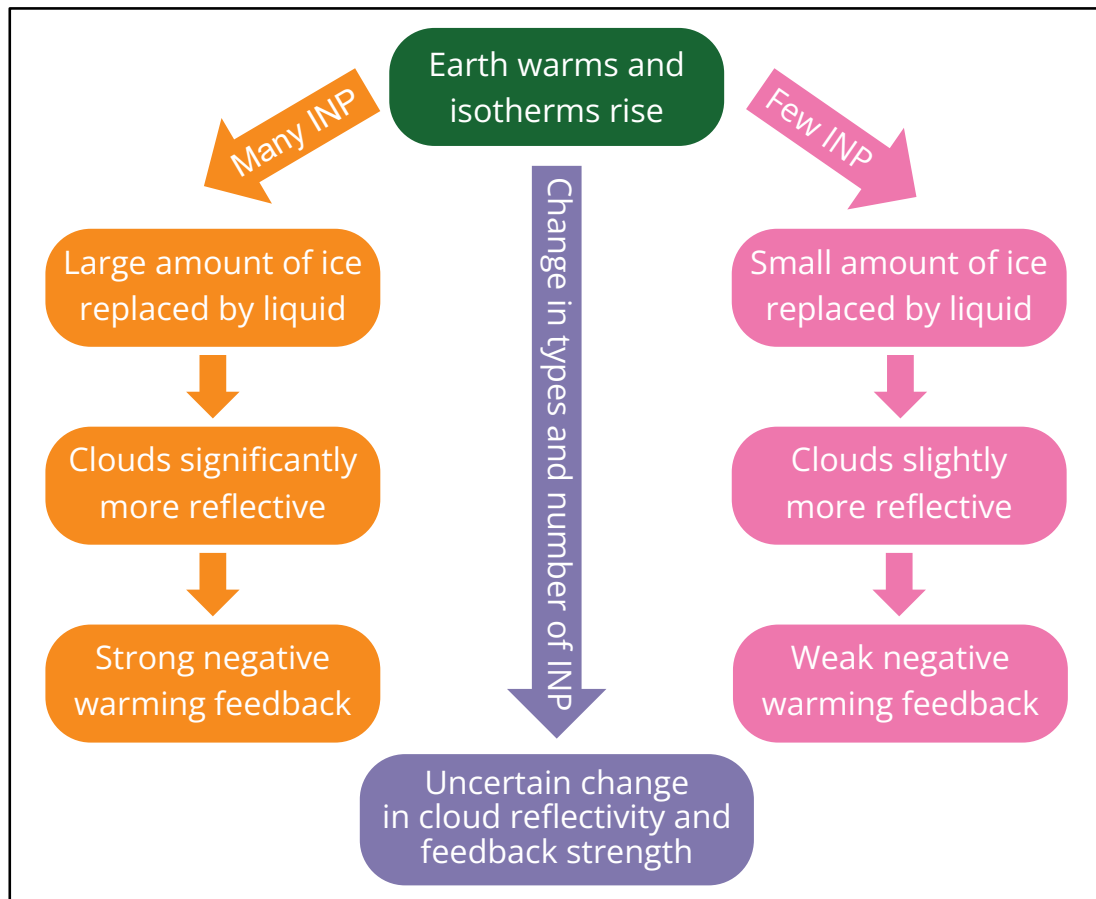


Figure 1.7: The shallow mixed-phase cloud feedback and how it is modulated by INPs, based on schematics from Storelvmo et al. (2015) and Murray et al. (2021).

increased dust emissions (Field et al., 2010), while biological INPs are likely to be released from melting permafrost (Creamean et al., 2020). Increased availability of INPs may reverse the direction of the cloud-phase feedback by increasing the relative concentration of ice in mixed-phase clouds (Murray et al., 2021). When testing this hypothesis in simulation using the CESM2 climate model, McGraw et al. (2023) identified this cloud feedback, while also finding that INPs affected the strength of other cloud feedbacks, though they found that aerosol and aerosol-unaware INP schemes produced cloud-phase feedbacks of similar magnitudes. While potentially important for cloud feedbacks, the INP-modulated changes in cloud albedo and cover are not thought to contribute significantly to radiative forcing since the most effective INPs in mixed-phase cloud regimes are not anthropogenic (Gettelman et al., 2012).

1.5 Cold-air outbreaks

Cold-air outbreaks (CAOs) are ejections of air from one of the polar regions to warmer lower latitudes (Brümmer, 1996; Vavrus et al., 2006; Kolstad and Bracegirdle, 2008; Kolstad et al.,

2009).² These happen over both land and marine regions, but when CAOs occur over marine regions, they are associated with large mixed-phase cloud decks which are susceptible to the influence of microphysical changes (Huang et al., 2025). Poor representations of marine CAOs have been shown to contribute to the radiation biases in global climate models (Bodas-Salcedo et al., 2012), and are representative of typical mixed-phase clouds influenced by the cloud phase feedbacks (Frey and Kay, 2018; Murray et al., 2021). Consequently, it is important to understand the behaviour of INPs in marine CAOs. Herein, the term CAO will be used to refer to marine CAOs, the focus of this thesis, unless otherwise stated.

CAOs occur frequently in the mid- to high-latitudes of both hemispheres and are most common in autumn, winter and spring, though they are not unheard of in summer, especially in the Southern Hemisphere (Fletcher et al., 2016a). In the Northern Hemisphere, intense CAOs are most common over the Labrador Sea and Fram Strait, the Norwegian and Barents seas (especially when air is sourced from Novaya Zemlya) and the Pacific Ocean east of Japan (Brümmer and Pohlmann, 2000; Fletcher et al., 2016a,b; Papritz and Spengler, 2017). In the Southern Hemisphere, CAOs occur at all longitudes north of Antarctica but are most intense in the Ross and Bellingshausen seas (Fletcher et al., 2016a).

1.5.1 Dynamics of cold-air outbreaks

Cold-air outbreaks typically occur when the prevailing wind is meridional and equatorward, often in the presence of a blocking system (Pithan et al., 2018; Nygård et al., 2023). Once the airmass of the CAO reaches the warmer marine surface, the large difference in temperature between the cold, relatively dry air of the CAO and the warmer ocean surface results in large fluxes of heat and moisture, driving convection (e.g. Brümmer, 1996; Renfrew and Moore, 1999). This convection takes the form of organised roll vortices, with stratocumulus “cloud streets” forming at the top of the layer (Etling and Brown, 1993). These distinctive cloud streets are characteristic of CAOs. Moving downstream, the CAO entrains dry air from above the stratocumulus layer (Deardorff, 1976) while the boundary layer deepens, causing the cloud layer to decouple from the surface and stratocumulus to dissipate in the absence of moisture supply (Sandu and Stevens, 2011; Murray-Watson et al., 2023). The CAO develops into open convective cells with cumulus clouds formed as convection is driven by the warming of the below-cloud layer by the ocean surface, while the stratocumulus dissipates having lost its source of moisture now the cloud layer is decoupled. This process is known as the stratocumulus-to-cumulus transition (SCT).

²Occasionally, the definition of CAO is extended to include relatively cold continental non-polar air which is advected towards a relatively warm surface (Fletcher et al., 2016a). Additionally, the term CAO is sometimes used to refer to extreme cold events, regardless of their underlying meteorological cause (e.g. Cellitti et al., 2006; Smith and Sheridan, 2020).

1.5.2 Microphysics of cold-air outbreaks and the role of INPs

While large-scale dynamics control the formation of a CAO, microphysical processes controlled by INPs can impact CAO cloud properties and the onset of the SCT. When exploring the sensitivity of a North Sea CAO to the concentration of ice, Abel et al. (2017) found that the ice concentration controlled the onset of precipitation, which in turn controlled the timing of the SCT by promoting the decoupling of the boundary layer. These findings were replicated by Wu et al. (2025) when simulating a Norwegian Sea CAO. Abel et al. (2017) also found a strong reduction in concentration of accumulation mode aerosol after the SCT, which suggests that precipitation acted to efficiently remove aerosol. de Roode et al. (2019) made a similar finding when modelling an East Atlantic CAO. In contrast to Abel et al. (2017), Tornow et al. (2021) found that the riming of ice early in a CAO accelerated the SCT of a West Atlantic CAO by reducing the amount of cloud water, depleting CCN and causing early light precipitation, though they found that the onset of heavier precipitation was not strongly controlled by ice. Neither Abel et al. (2017) nor de Roode et al. (2019) simulated INPs, while INPs in Tornow et al. (2021) were temperature independent and diagnostically maintained a pre-determined ice particle concentration, effectively representing ice particle concentration rather than ice nucleation (Ovchinnikov et al., 2014). To test the response of CAO clouds to ice nucleation, Huang et al. (2025) perturbed the magnitude of ice produced when modelling two Labrador Sea CAOs using a temperature-dependent INP parameterisation (Cooper, 1986), while also perturbing the efficiency of the HM process and the background droplet number concentration. In both warm and cold CAOs, changes to the INP concentration had the largest changes to bulk cloud properties including liquid and ice water path.

1.5.3 Metrics to define a CAO

The presence and strength of a CAO is often measured using variants of the cold-air outbreak index, M , on a specific vertical pressure level p . In recent studies, this is now conventionally defined as

$$M_p = \theta_{\text{skt}} - \theta_p \quad (1.17)$$

where θ_p is the potential temperature at the chosen pressure level and θ_{skt} is the surface skin potential temperature (e.g. Fletcher et al., 2016a,b; Terpstra et al., 2021). However, earlier works used variants of this with different normalisations (Kolstad and Bracegirdle, 2008; Kolstad et al., 2009; Bracegirdle and Kolstad, 2010). Regions where $M_p > 0$ are considered to be a CAO, while greater values of M_p indicate the CAO is stronger (Papritz and Spengler, 2017). While the form of M_p is largely consistent between studies, many different pressure levels have been used to prioritise different characteristics of CAOs when establishing their strength. Papritz et al. (2015) suggested that the original choice of $p = 700$ hPa by Kolstad and Bracegirdle (2008) best captures the vertical static stability of an outbreak but chose $p = 850$ hPa in their study to capture the intensity of heat fluxes, while noting that levels as high as 500 hPa have been used. The choice of p also affects the number of

CAOs detected in climatologies, with Fletcher et al. (2016a) finding that $p = 800$ hPa biased towards the detection of high-latitude CAOs when compared to $p = 700$ hPa, while choosing $p = 850$ hPa biased towards the detection of subtropical CAOs. However, this measure is imperfect, as observations of typical CAO conditions such as convective rolls of cloud streets following a meridional wind have occurred in regions where $M < 0$ (Clarke et al., 2025, *submitted*). Additionally, the nature of the CAO index means that $M < 0$ over sea ice, so it fails to identify CAOs before the development of the cloud system. While CAO index is the most commonly used metric, Terpstra et al. (2021) defined an alternative metric, the cold-air outbreak depth, H , to measure the spatial extent of CAOs.

$$H = p_{\theta=\theta_{\text{skt}}} - p_{\text{surf}}, \quad (1.18)$$

where $p_{\theta=\theta_{\text{skt}}}$ is the pressure level which has potential temperature equal to θ_{skt} and p_{surf} is the surface pressure. This was able to better capture extreme CAO events.

1.5.4 Observations of cold-air outbreaks

Given their high frequency of occurrence and relatively consistent morphology, Murray et al. (2025, *submitted*) have described CAOs as a “natural laboratory” for in-situ observations of mixed-phase cloud physics. This is because the formation of cloud streets is typically pinned to the outflow over land or sea ice (Fletcher et al., 2016a), which means flight campaigns can be planned around a predictable formation location given knowledge of the synoptic conditions (Murray et al., 2025, *submitted*).

Indeed, there have now been over eighty years of in-situ study of CAOs, starting with the analysis of sonde measurements by Lettau (1944). Approximately fifteen years later, Bunker (1960) made the first airborne measurements of CAOs in a study of heat fluxes in different latitudes and seasons. In November 1970, Lenschow (1973) and LeMone (1973) performed and analysed the first airborne measurements targeted at CAO clouds, making measurements of water mixing ratio, heat and energy fluxes in CAOs over the Great Lakes to better understand the thermodynamics of CAOs and structure of their convective rolls. The 1970s and 1980s saw many small-scale observational experiments of CAOs (e.g. Ninomiya and Akiyama, 1976; Brümmer et al., 1985) before the ARKTIS campaign in 1993 made the first large-scale experiment to make measurements of an entire CAO system over the Norwegian Sea (Brümmer, 1996). Unlike previous campaigns, ARKTIS co-ordinated measurements between multiple aircraft, ships and radiosonde stations from the initial ejection of the air-mass from the sea ice to the initial transition into cumulus. However, the measurements made were largely dynamical and focused on bulk measurements of CAO properties (Brümmer, 1996). Renfrew and Moore (1999) were able to make in-situ observations deeper into the cumulus regime during the Labrador Sea Deep Convection Experiment of 1997, again focusing on dynamical measurements to understand the structure of the roll vortices and heat fluxes.

In contrast to dynamical measurements, in-situ measurements of microphysics in CAOs are relatively recent. Some polar aircraft campaigns in the mid-2010s, such as the ACCACIA campaign (Young et al., 2016; Abel et al., 2017) and the Southern Ocean Cloud Radiation and Aerosol Transport Experimental Study (SOCRATES) observed CAO conditions while measuring microphysical quantities such hydrometeor and aerosol concentrations. However, neither of these campaigns systematically targeted CAOs.

The first CAO-targeted field campaign to make a suite of microphysical measurements was the 2018 Iceland-Greenland Seas Project (IGP), which had size spectra measurements of aerosol and liquid droplets, but no measurements of ice (Renfrew et al., 2019), since the campaign was focused on the ocean response to CAOs (Renfrew et al., 2023). The 2019-22 Aerosol Cloud meteorology Interactions over the western ATLantic Experiment (ACTIVATE) off the east coast of the United States added ice concentration measurements to their suite of microphysical measurements.

However, it was not until the 2022 Arctic Cold Air Outbreak (ACAO) campaign over the Norwegian and Barents seas (discussed at length in Section 2.2.1) that systematic airborne sampling of INPs was performed in addition to measurements of ice and liquid (Raif et al., 2024), though systematic measurements of INPs in CAOs were made during the ground-based Cold-air Outbreaks in the Marine Boundary Layer Experiment (COMBLE) (Geerts et al., 2022). Since the ACAO campaign, there have been further campaigns with similar instrumental capabilities, including the Cold-Air Outbreaks over the Sub-Arctic Region (CAESAR) campaign which took place in 2024 over the Norwegian and Greenland seas (Zuidema et al., 2025), the 2022 M-Phase experiment over the Labrador Sea (Huang et al., 2025) and the 2023 Southern Ocean Clouds experiment off the Antarctic Peninsula (Smith et al., 2025).

1.6 Objectives

This thesis will use the natural laboratory of cold-air outbreaks to investigate aerosol and INP properties, as well as the effects of INPs on high-latitude mixed-phase clouds.

In Chapter 2, I present measurements of ice-nucleating particles in cold-air outbreaks made during an aircraft-based field campaign to answer the following questions:

- What concentrations of INPs are found in Arctic CAOs?
- What are the sources of INPs in Arctic cold-air outbreaks?

To further investigate the behaviour of INPs in CAOs, I perform computer modelling using Cloud AeroSol Interacting Microphysics (CASIM), a two-moment microphysics model. The processes of CASIM, its coupling to the Met Office Unified Model and its ability to represent mixed-phase clouds is described in Chapter 3.

In Chapter 4, I present modelling work using a case study from Chapter 2 which showed

evidence of INP processing and removal. Here, I answer the questions:

- How well can a two-moment microphysics scheme with active INPs capture observed variations of INP concentration in a cold-air outbreak system?
- What drives INP removal in a cold-air outbreak?

In Chapter 5, I build on the modelling work in Chapter 4 by implementing an INP scheme makes a different assumption about the distribution of INPs among aerosol. Here, I answer the question:

- Can the removal of INPs be better-represented in a regional model using an INP scheme which limits ice nucleation according to the number of INPs rather than assuming that there is a constant relationship between the number of INPs and the aerosol concentration?

Finally, in Chapter 6, I synthesise conclusions from each of Chapters 2, 4 and 5 to make recommendations for future research.

Chapter 2

High ice-nucleating particle concentrations associated with Arctic haze in springtime cold-air outbreaks

Abstract

The global variation in ice-nucleating particle (INP) concentrations is an important modulator of the cloud-phase feedback, where the albedo of mixed-phase clouds increases in a warming climate. Shallow clouds, such as those observed in cold-air outbreaks (CAOs), are particularly important for cloud-phase feedbacks and highly sensitive to INPs. To investigate the sources and concentrations of INPs in CAOs, we made airborne measurements over the Norwegian and Barents seas as part of the March 2022 Arctic Cold-Air Outbreak (ACAO) field campaign. Aerosol samples were collected on filters at locations above, below and upstream of CAO cloud decks. Throughout the campaign, INP concentrations were comparable to the highest concentrations previously observed in the Arctic. Scanning electron microscopy analysis of samples taken upstream of cloud decks showed that supermicron aerosol was dominated by mineral dusts. Analysis of aerosol particle size measurements to obtain an INP active site density suggested sea spray was unlikely to be the dominant INP type. These site densities were also too great for mineral components alone to be the dominant INP type above -20°C . Accordingly, it is likely that the dominant INP type was mineral dust mixed with other ice-nucleating materials, possibly of biogenic origin. Back-trajectory analysis and meteorological conditions suggested a lack of local INP sources. We therefore hypothesise that the high INP concentration is most likely to be associated with aged aerosol in Arctic haze that has undergone long-range transport from lower-latitude regions.

2.1 Introduction

As discussed in Chapter 1, the micro- and macrophysics of CAO cloud decks are susceptible to the influence of INPs, while the INP-moderated cloud-phase feedback is likely to be particularly sensitive to CAO clouds due to their prevalence and strong contrast in albedo with the ocean beneath. However, there is substantial uncertainty over the INP concentrations within CAOs, which makes it difficult to assess their effects.

Globally, INP concentrations vary substantially both spatially and temporally, but some of the greatest variability observed anywhere on Earth occurs in the Arctic, where CAOs abound. Measurements of INPs in the Arctic reveal some of the lowest INP concentrations on Earth, comparable with the Southern Ocean, and some of the greatest, comparable with mid-latitude locations where terrestrial biological INP types dominate (Murray et al., 2021). Variability occurs both on seasonal scales (Wex et al., 2019; Sze et al., 2023) and strongly on timescales of hours to days (Porter et al., 2022). This indicates not only that the sources of INPs within and transported to the Arctic are variable, but also that removal processes are important, since periods of high INP concentration can be followed by periods of low INP concentration.

Figure 2.1a shows the potential sources of INPs for CAO clouds in the Northern Hemisphere. Potential Arctic sources of INPs are biogenic material from sea-spray (Wilson et al., 2015; DeMott et al., 2016; McCluskey et al., 2018b) and terrestrial INPs from high-latitude sources (Kawai et al., 2023). Potential terrestrial sources of INPs include glacial outwash sediments (Tobo et al., 2019; Xi et al., 2022; Barr et al., 2023), deserts in Iceland (Sanchez-Marroquin et al., 2020), boreal forests (Brasseur et al., 2022), Arctic surface vegetation (Pereira Freitas et al., 2023) and thawing permafrost (Creamean et al., 2020). In addition, Arctic aerosol can have lifetimes of many weeks, which results in the build up of Arctic haze in winter and spring (Stohl, 2006). Accordingly, it is also possible that INPs entering CAOs may have been transported into the Arctic from lower latitudes, residing in the stable Arctic atmosphere before being transported out again in CAOs (Carslaw, 2022b). Different types of INPs have different characteristic ranges of freezing temperatures (Kanji et al., 2017). This means understanding the aerosol sources that contribute to the INP population in CAO clouds can reduce the uncertainty in the cloud-phase feedback.

To understand the sources of INPs in CAO clouds, there is a need for localised airborne aerosol measurements to capture INP concentrations throughout a CAO, especially in the Northern Hemisphere. As discussed at length in Section 1.5.4, Northern Hemisphere aircraft campaigns have characterised the dynamics of cold-air outbreaks (Brümmer, 1996) over the Norwegian Sea and have made airborne measurements of aerosol and clouds without INP measurements in mid-latitude cold-air outbreak conditions (Young et al., 2016; Sorooshian et al., 2019). INP measurements have been made in CAOs from the ground in the Northern Hemisphere (Geerts et al., 2022; Gjelsvik et al., 2025) and from aircraft in

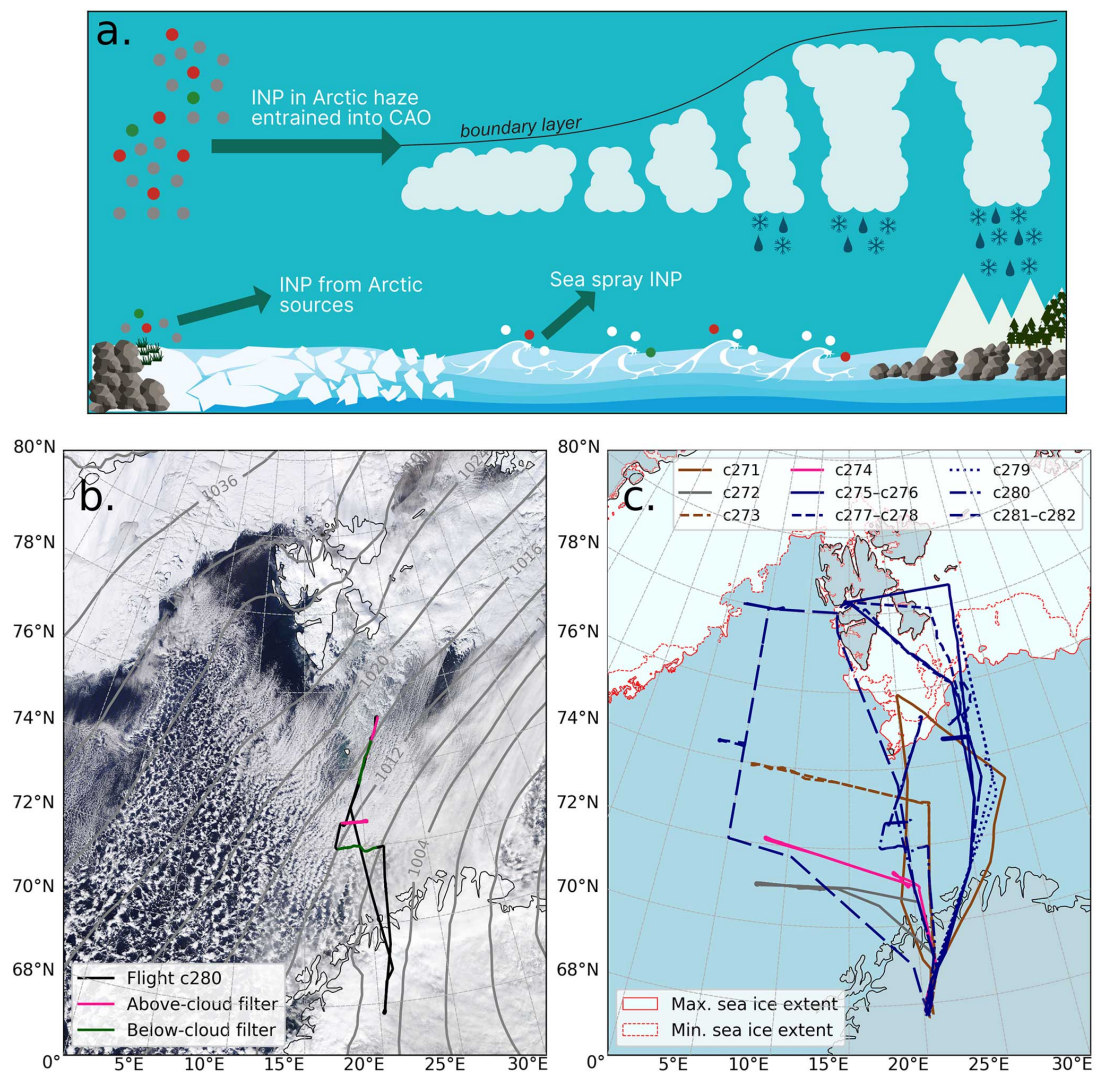


Figure 2.1: Panel (a) shows a schematic representation of an Arctic cold-air outbreak and potential sources of aerosol to the cloud system. The green and red circles depict biological and abiological INPs respectively, while the grey and white circles depict aerosols that do not nucleate ice in CAOs. Panel (b) shows satellite imagery taken on 29 March 2022 by the Moderate Resolution Imaging Spectroradiometer (MODIS) of a typical CAO measured during the A CAO campaign (MODIS Land Science Team, 2020). The path of flight c280, which probed this CAO, is overlaid along with the locations of INP filter samples, which were taken between 30 min and 2 h before the image time. Sea-level pressure isobars from ERA5 reanalysis are also overlaid (C3S, 2018). Panel (c) shows the flight paths from all 9 flying days during the campaign. The blue, pink and brown flight paths represent CAOs with predominantly N, NW and W flows respectively. Flight c272 probed a warm-air intrusion and is coloured grey. Sea ice extent in this and other maps was obtained from the Multisensor Analyzed Sea Ice Extent (MASIE) dataset (U.S. National Ice Center et al., 2010). The flight track for flight c278 is incomplete due to equipment failure.

the Southern Hemisphere (McFarquhar et al., 2021; McCluskey et al., 2018a). In this study, we focus on cold-air outbreaks over the Norwegian and Barents seas to report the first airborne measurements of INPs in Northern Hemisphere cold-air outbreaks. Having reported these measurements, we characterise the INP concentrations observed and try to identify the primary INP source(s) through a process of elimination of potential sources.

2.2 Methodology

2.2.1 The Arctic Cold-Air Outbreak campaign

The Arctic Cold-Air Outbreak (ACAO) flight campaign took place over the Norwegian and Barents seas in March 2022. The campaign used the BAe 146 aircraft of the Facility for Airborne Atmospheric Measurements (FAAM) to make measurements in cold-air outbreaks. A key aim was to investigate the development and evolution of the cloud systems and the associated aerosol and cloud microphysical properties. A total of 12 flights were performed on nine days between 11 and 30 March 2022. The tracks of these flights are depicted in Figure 2.1c. Where possible, flight days and locations were chosen to capture CAOs with characteristics such as cloud streets that were clearly positioned away from fronts to ensure well-defined air flows and air masses. CAO conditions, of varying strength, occurred on 8 of the 9 flying days. CAO index values, calculated according to Eq. 1.17 using 800 hPa as the reference pressure level, are shown in Section A.1. On the other day (flight c272), a warm-air intrusion (an injection of mid-latitude air into the Arctic; Pithan et al., 2018) occurred and was measured as a contrasting case. Flights c271 and c273 probed CAOs with westerly flows with air flowing off Greenland, while the flow during c274 was northwesterly. Flights c275 to c282 probed CAOs with northerly or north by northeasterly flows. Aerosol measurements were made online (i.e. continuously) using underwing optical probes, and filter samples were collected for specific periods for subsequent offline analysis.

2.2.2 Filter samples for INP analysis

Filter samples were collected using the aircraft inlet system, previously characterised by Sanchez-Marroquin et al. (2019). In this system, two parallel inlets are aligned with the heading of the aircraft, and the airflow through each inlet is assisted by a vacuum pump. The inlet system is designed to minimise turbulence and flow separation when flying at a low Mach number (Andreae et al., 1988, 2000) and has a bypass to remove droplets and ice crystals (Talbot et al., 1990). The bypass also increases the flow rate, reducing the sub-isokinetic enhancement of larger particles (Sanchez-Marroquin et al., 2019). A different type of filter was used in each inlet so that different types of analyses could be performed on aerosol samples from the same location and time. Polytetrafluoroethylene (PTFE) membrane filters were used for INP concentration measurements via droplet-freezing assays, while polycarbonate track-etched membrane filters were used for scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS) aerosol composition analysis.

Air entering each inlet passes through a sampling line into a filter holder previously described by Price et al. (2018) and Sanchez-Marroquin et al. (2019). Air that passes through the filter holder is measured by a mass flow meter, which reports the flow in standard litres (273.15 K, 101.325 kPa). Inside the filter holder, the filter sits on a stainless steel support mesh. The PTFE filter (Sartorius, product number 11803-47-N) has a pore size of 1.2 μm ,

while the polycarbonate filter (Whatman, product number 111107) has a pore size of $0.4\ \mu\text{m}$. The PTFE filter allowed more throughput for INP analysis, while the polycarbonate filter had less throughput. Two types of steel mesh were used during the campaign – one manufactured by Millipore (product number XX4304707) and one manufactured by MesaLabs (product number 1584). Using the MesaLabs meshes enhanced the flow through the filter, though flow rates were consistently greater through PTFE filters than through polycarbonate filters. Flow through the filter was also dependent on altitude, with higher flow rates at lower altitudes. The largest volume of air sampled by a PTFE filter was $(1730 \pm 30)\text{ L}$, while the smallest volume of air sampled was $(473 \pm 9)\text{ L}$. For polycarbonate filters, the largest volume sampled was $(990 \pm 20)\text{ L}$, while the smallest was $(276 \pm 6)\text{ L}$. There was no correlation between the INP concentrations measured and the volume of air sampled by each filter. Handling blanks were collected to determine the baseline ice-nucleating activity of the procedure and equipment used. These handling blanks were collected during the flight by loading filters as normal into the aircraft filter holder system, opening the filter system up to air flow for up to 2 s, then closing the filter system and removing the filters as normal. Handling blank filters were processed for INP analysis in the same manner as the aerosol sample filters.

Figure 2.2a shows the location of the filter sample measurements over the Norwegian and Barents seas between 71°N to 79°N . The altitudes of filter measurements were between 40 m and 3400 m, and all samples were taken in air with no clouds or precipitation. The majority of samples were taken above or below the CAO clouds, although two samples (c276r2 and c278r2) were taken upstream of the cloud onset. Sampling periods were typically between 18 min and 28 min, excluding pauses in sampling where the inlets were temporarily closed when the aircraft turned or flew through precipitation. Only one of the samples (c273r2) had a shorter sampling time of 14 min, while one of the samples (c273r1) had a longer sampling time of 37 min. However, both of these filters sampled volumes of air that were not at the extremities of the volume range sampled.

To derive INP concentrations from the samples, droplet-freezing assays were performed according to a drop-on technique described fully by Price et al. (2018), which makes use of the microlitre Nucleation by Immersed Particle Instrument (Whale et al., 2015). Filters were processed in a field laboratory set up in a hotel room to minimise the storage and transport effects that are known to lead to loss of INPs (Beall et al., 2020). All but one of the filters were processed within 24 h of collection. In these experiments, rectangular glass cover slips ($48\text{ mm} \times 64\text{ mm}$, no. 1.5 thickness, Agar Scientific, UK) were washed with purified water (high-performance liquid chromatography grade, Sigma-Aldrich, UK) and isopropanol (VWR, UK) and then immersed for 1 h in a petri dish containing Turtle Wax ClearVue rain repellent solution (Rapid Electronics, UK) to render them hydrophobic. The slides were removed from the ClearVue solution, rinsed with isopropanol and water, and then dried with particle-free air from an oil-free air compressor (24 L, model HY7524, Hyundai, UK, with a HEPA filter in line). In a laminar flow hood, a filter onto which aerosol had been

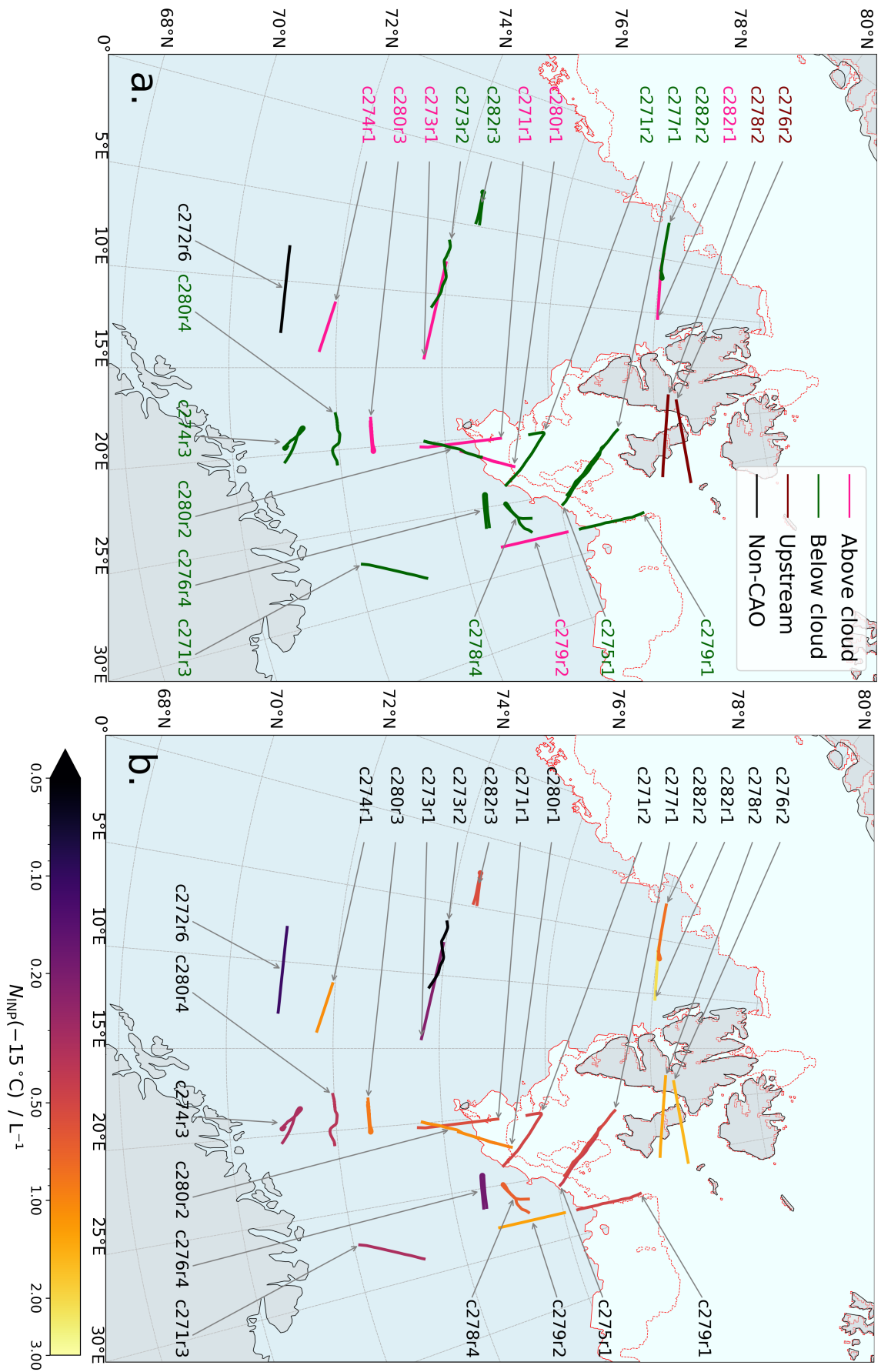


Figure 2.2: Locations of all filter samples taken during the ACAO campaign. Panel (a) displays the location of the sample relative to the cloud deck, while panel (b) shows $M_{\text{INP}}(-15^\circ\text{C})$, the INP concentration at -15°C for each sample. All filters except c272r6 were taken in CAO conditions.

sampled was placed onto a hydrophobic slide, and between 60 and 70 droplets of purified water with a volume of 2 μL were pipetted directly onto the filter using a manual electronic pipette. The hydrophobic glass slide-filter assembly was then placed onto a cold plate of an Asymptote EF600 Stirling engine-based cold stage with a drop of silicone oil in between to aid thermal contact. A flow of HEPA-filtered compressed air that had been passed through a drying column (Drierite 26800, Sigma-Aldrich, UK) was used to flush the volume above the droplets to prevent frost formation. The filter was then cooled at a rate of 1 K min^{-1} until all droplets froze. The cooling was recorded by a camera, and the temperature of the cold plate was measured concurrently. Software written in Python was used to automatically detect freezing events and determine the temperature at which each droplet froze (Barr, 2023). The same procedure was used for handling blanks from the aircraft. In addition, filter blanks were run each day, consisting of purified water droplets pipetted onto new, untouched filters to ensure that the procedure and equipment were operating correctly and without unacceptable contamination (or that the system was cleaned and procedures were improved until contamination levels were deemed acceptable).

An alternative assay technique is to wash aerosol material off the filters in a larger volume of deionised water and take microlitre droplets from this volume for freezing experiments (O'Sullivan et al., 2018). Given the relatively short collection time for aerosols on the filters, the drop-on technique was chosen because it utilises a greater fraction of aerosols collected during the sampling period (approximately 30 times more aerosol per droplet). This gives greater sensitivity to the relatively small fraction of all aerosols that are able to nucleate ice (Sanchez-Marroquin et al., 2021). Using filters collected in parallel, comparison between the wash-off and drop-on techniques shows good agreement in the INP concentration ranges where the techniques overlap (Sanchez-Marroquin et al., 2021). However, in one-third of cases compared by Sanchez-Marroquin et al. (2021), the wash-off technique produced lower INP concentrations at temperatures above -20°C , temperatures at which biological and biologically enhanced INPs are active. The wash-off technique agrees well with other online and offline INP analysis techniques for mineral dusts (DeMott et al., 2018) but produced lower INP concentrations for a cellulose sample compared to other techniques (Hiranuma et al., 2019). Similarly, the wash-off technique produced lower INP concentrations than a liquid impinger technique when measuring INPs in the Arctic, which was attributed to the larger size cut-off of the impinger technique (Li et al., 2023).

From droplet-freezing temperatures, INP temperature spectra were derived using the framework of Vali (1971, 2019). For each assay, freezing events were grouped into temperature bins with a size of $\Delta T = 1 \text{ K}$ to calculate the differential nucleus spectrum,

$$k_{\text{assay}}(T) = -\frac{1}{V_d \Delta T} \ln \left(1 - \frac{\Delta N_f}{N_f(T)} \right), \quad (2.1)$$

where V_d is the droplet volume (2 μL), ΔN_f is the number of droplets that freeze in a temperature interval and $N_f(T)$ is the number of droplets unfrozen at T , the upper limit of

the bin's temperature range. The error in ΔN_f according to the Poisson distribution was propagated to find the error in $k_{\text{assay}}(T)$ for the individual spectrum. The average differential INP spectrum of the background filters, k_{bg} , was subtracted from the spectra for an individual assay to get the "true" differential INP spectrum for a sample,

$$k(T) = k_{\text{assay}}(T) - k_{\text{bg}}(T). \quad (2.2)$$

From this, the INP concentration, N_{INP} , was calculated as

$$N_{\text{INP}}(T) = \frac{V_d A_f}{V_a A_d} \Delta T \sum_0^T k(T), \quad (2.3)$$

where V_a was the volume of air sampled; A_f was the surface area of the filter exposed to aerosol, $(11 \pm 2) \text{ cm}^2$; and A_d was the surface area of the filter in contact with a droplet, $(0.014 \pm 0.002) \text{ cm}^2$. A_d was calculated using spherical cap geometry with a contact angle of 126° as in Price et al. (2018).

2.2.3 Scanning electron microscopy

Using the method described by Sanchez-Marroquin et al. (2019), SEM-EDS was used on two filters to ascertain the size-resolved elemental composition of aerosol captured on the polycarbonate filters. The elemental composition was used to estimate the fraction of aerosol from terrestrial dust and marine sources. Portions of the filters were mounted on 25 mm diameter stubs, sputter-coated with 30 nm of platinum and analysed at the University of Leeds with a Tescan Vega3 XM electron microscope fitted with an Oxford Instruments X-Max 150 SDD (silicon drift detector) energy-dispersive X-ray spectrometer. Using AZtec 3.3 feature recognition software, the filters were scanned for aerosol particles, and a prescribed algorithm was used to automatically classify particles into categories (e.g. mineral dust and sea spray) using their elemental composition. This classification algorithm is further described in Sanchez-Marroquin et al. (2019). The carbonaceous category of particles includes particles which contain no elements other than O and C and therefore does not distinguish between organic and elemental carbon. Biological particles are therefore not explicitly classified using this method but can be qualitatively identified on the basis of size and morphology (Sanchez-Marroquin et al., 2021). The classified particles were then binned according to their equivalent spherical diameter. A minimum diameter of $0.3 \mu\text{m}$ was chosen as it is comfortably above the size threshold at which the feature recognition software erroneously detects features. The total surface area of aerosol sampled in each composition category was normalised by the fraction of filter area analysed and the volume of air sampled during the sampling run.

2.2.4 Optical probes

Two underwing optical probes were used to measure the aerosol number, surface area and volume concentrations during the flight. These were the Passive Cavity Aerosol Spectrometer Probe (PCASP) 100X (Droplet Measurement Technologies with SPP200 electronics) and Cloud Droplet Probe (CDP; Droplet Measurement Technologies). Both probes operate by measuring the intensity of radiation from a laser that is scattered by incident particles. For the CDP, sampled particles pass directly through the beam, while for the PCASP, a sample is enclosed in clean air before entering an optical chamber. Both probes use the magnitude of the scattered radiation to assign the particle to 1 of 30 bins associated with the diameter of the particle (Rosenberg et al., 2012). Although the CDP is designed to measure droplets, it has previously been used to measure coarse-mode aerosol by adjusting the calibration of the probe to assume a refractive index representative of the aerosol sampled (Sanchez-Marroquin et al., 2020). To do this, the calibration method of Rosenberg et al. (2012) was used to calculate the particle diameters measured by each bin when given a refractive index. For this study, particle size distributions were first obtained using real refractive indices ranging from 1.5 to 1.7 and imaginary components ranging from $0i$ to $0.003i$ as in Sanchez-Marroquin et al. (2020). Since the error associated with the refractive index was small, a refractive index value of $1.56 + 0i$ was chosen for consistency with Sanchez-Marroquin et al. (2020). When using a refractive index of 1.56, the PCASP measures particles with a diameter range of $0.1\ \mu\text{m}$ to $3\ \mu\text{m}$, while the CDP measures particles with a diameter range of $4.5\ \mu\text{m}$ to $70\ \mu\text{m}$.

Previous studies have found that the coarse-mode aerosol number reported by the PCASP and CDP probes is suppressed relative to particles counted on filter samples using SEM (Sanchez-Marroquin et al., 2019; Barr, 2023). The use of larger-pore filters and a fully open bypass line (as described in Section 2.2.2) minimised this discrepancy.

To ensure that no cloud droplets or swollen hygroscopic aerosol was reported by the CDP, CDP data have only been used when relative humidity (RH) was below 80 % and when cloud-free air was identified using the Nevzorov hot-wire instrument. The Nevzorov instrument derives the liquid and total condensed water content (liquid and ice) based on the power required to melt and evaporate hydrometeors collected by the instrument (Korolev et al., 1998; Abel et al., 2014). The power supplied to the Nevzorov probe is significantly different in and out of the cloud. Using this variation in power supply, a 1 Hz cloud flag was derived to partition the data as in or out of the cloud in a manner similar to Barrett et al. (2020).

2.3 Results

Successful INP analysis was performed on 23 filter samples. All but filter c272r6 were taken when mixed-phase boundary layer clouds associated with cold-air outbreaks were present. A total of 13 samples were taken above the cloud deck, 8 were taken below the cloud deck

and 2 were taken upstream of the development of the cloud in clear air. These sampling locations were considered to represent air below, above and before the development of the atmospheric boundary layer over sea respectively. The locations, altitudes and purpose of each filter measurement are shown in Table 2.1. Figure 2.2a shows that the filter samples were distributed throughout the development of the cold-air outbreaks.

2.3.1 INP concentration spectra

Figure 2.3a shows a composite of the INP concentration spectra, $N_{\text{INP}}(T)$, measured during the campaign. Measurements on and after 21 March (c274 onward, when the flows had a northerly component) form a tight band of INP spectra, all falling within about 1 order of magnitude and centred at about 1 L^{-1} at -15°C . Spectra from 11 March through to 19 March (flights c271 to c273), when the flow directions were from the west or south, had much more variable INP concentrations, with one filter measurement recording an INP concentration below 0.1 L^{-1} at -15°C (c273r2) (see Figure A.2). The median CAO INP concentration during the campaign, $\bar{N}_{\text{INP}}$, was described by the function

$$\bar{N}_{\text{INP}} = \bar{a} \exp \left[\bar{b} (\bar{c} - T)^{\bar{d}} \right], \quad (2.4)$$

with fitting parameters $\bar{a} = 1.480 \times 10^{-2} \text{ L}^{-1}$, $\bar{b} = 1.271 \text{ K}^{-0.5}$, $\bar{c} = 226.2 \text{ K}$ and $\bar{d} = 0.5$. The fitting procedure used to obtain $\bar{N}_{\text{INP}}$ is described in Section A.3, along with similar four-parameter fits for each individual filter measurement. Figure 2.4 shows that INP concentrations during the ACAO campaign were among the highest recorded in the high latitudes of the Northern Hemisphere in springtime. They are comparable with the highest mid-latitude INP measurements collated by Petters and Wright (2015) and INP concentrations measured by Sanchez-Marroquin et al. (2020) in autumnal Icelandic dust plumes. They were also similar to the highest INP concentrations reported by Porter et al. (2022) at the North Pole in August 2018 when aerosol was transported from the Russian coast. Both Rogers et al. (2001) and Prenni et al. (2007) used an online thermal gradient diffusion chamber to quantify INPs from an aircraft north of Alaska during May and September–October respectively. While they often saw no detectable INPs, they also reported periods when INP concentrations were comparable to (or sometimes greater than) those found in ACAO. Sanchez-Marroquin et al. (2023) reported airborne INP measurements from March in northern Alaska using the same filter technique as used in ACAO, revealing INP concentrations at least 1–2 orders of magnitude lower than in ACAO, with a steeper slope (many measurements were consistent with the background). These flights were not focused on CAO events.

The only previous airborne springtime INP measurements made in the vicinity of Svalbard were made by Hartmann et al. (2020), who flew over sea ice in the Arctic Ocean and Fram Strait in late March and early April 2018 and collected aerosol onto polycarbonate filters for a wash-off droplet-freezing assay. Hartmann et al. (2020) collected filter samples from an entire flight rather than in specific locations and reported INP concentrations that

Table 2.1: Times, locations and sampling intent of all filter samples made during the campaign.

Sample ID	Run type	Date (mm-dd)	Start (UTC)	End (UTC)	Mean z / m	Mean lat. / °N	Mean lon. / °E	CAO flow	Notes
c271r1	above cloud	03-11	09:40:07	10:07:24	1747	74.3	20.3	W	
c271r2	below cloud	03-11	10:18:03	10:42:48	39	75.5	21.5	W	2 min pause during aircraft turning
c271r3	below cloud	03-11	11:18:05	11:38:38	49	72.7	27.9	W	filter processed 3 d after collection two pauses for precipitation
c272r6	above cloud	03-16	12:07:25	12:34:15	2957	71.0	10.5	N/A	warm-air intrusion conditions
c273r1	above cloud	03-19	12:01:25	12:38:51	3403	73.8	11.2	W	
c273r2	below cloud	03-19	12:54:07	13:13:35	415	73.9	8.7	W	two short pauses to avoid showers
c274r1	above cloud	03-21	13:47:10	14:07:23	2672	71.8	12.6	NW	
c274r3	below cloud	03-21	15:18:08	15:41:22	165	71.2	19.3	NW	two short pauses for aircraft turning and one pause to avoid showers
c275r1	below cloud	03-23	10:03:37	10:28:22	162	76.5	23.8	N	
c276r2	upstream	03-23	12:55:29	13:20:29	1711	78.5	22.0	N	
c276r4	below cloud	03-23	14:32:27	14:55:27	156	74.6	25.2	N	
c277r1	below cloud	03-24	10:30:52	10:59:06	159	76.7	22.8	N	two pauses for altitude increases (poor visibility) cloud was in the very early stages of formation
c278r2	upstream	03-24	13:02:05	13:26:16	1725	78.2	21.3	N	
c278r4	below cloud	03-24	14:07:00	14:31:56	333	75.1	26.0	N	2 min pause for aircraft turning due to precip.
c279r1	below cloud	03-25	11:23:21	11:44:12	155	76.9	28.0	N	
c279r2	above cloud	03-25	11:48:26	12:08:44	1323	75.4	28.0	N	
c280r1	above cloud	03-29	09:29:00	09:49:17	1573	74.8	21.6	N	
c280r2	below cloud	03-29	09:59:55	10:15:58	327	74.1	20.6	N	
c280r3	above cloud	03-29	10:34:01	10:54:06	2013	72.6	19.4	N	2 min pause during aircraft turning
c280r4	below cloud	03-29	11:05:35	11:24:51	334	72.0	19.3	N	1 min pause to avoid showers
c282r1	above cloud	03-30	12:08:32	12:28:27	1761	78.0	7.5	N	
c282r2	below cloud	03-30	12:40:23	13:01:59	308	78.0	4.6	N	brief pause for aircraft manoeuvre; the polycarbonate filter holder popped open so the filter was replaced after 5 min
c282r3	below cloud	03-30	14:02:12	14:26:08	307	74.4	3.5	N	4 min pause during aircraft turning

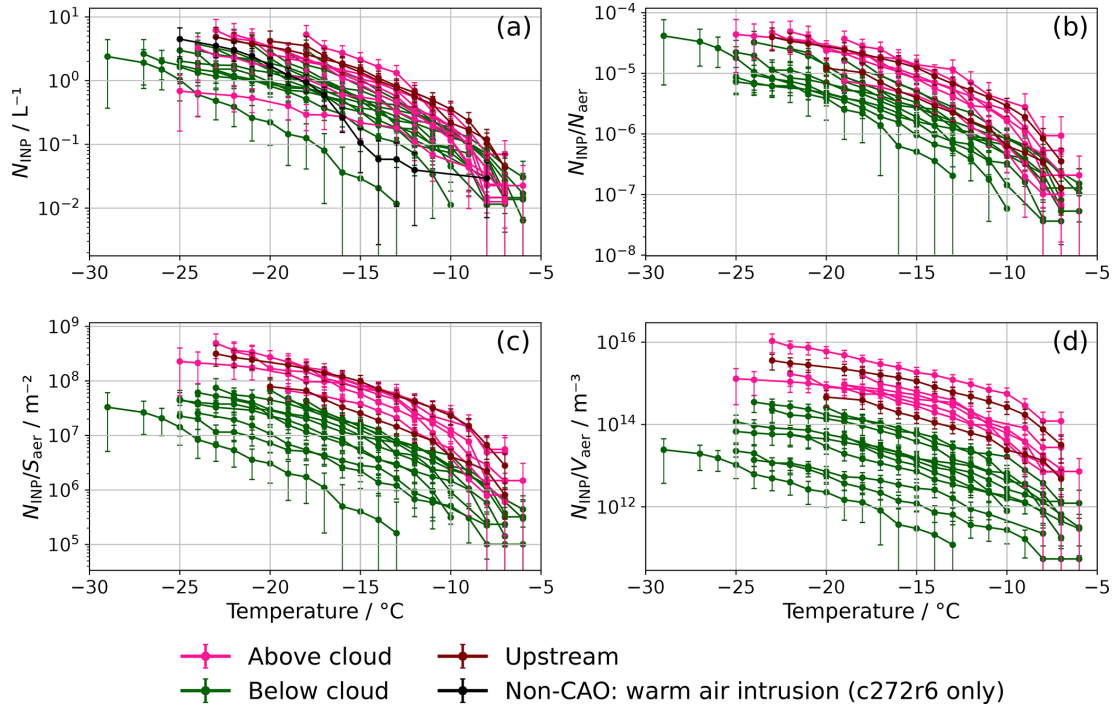


Figure 2.3: Panel (a) shows INP concentrations for each sample made during the campaign. Panels (b), (c) and (d) show these concentrations normalised by the total number (N_{aer}), surface area (S_{aer}) and volume (V_{aer}) of aerosols with diameters greater than $0.1 \mu\text{m}$. A version of this figure where each sample is individually labelled can be found in Section A.2.

were 1–2 orders of magnitude lower than those observed during ACAO in three out of nine samples. The other six samples had lower INP concentrations, often consistent with their handling blanks. The three higher INP samples, which contained heat-sensitive INPs, were associated with airflows with a strong northerly component, and satellite imagery (MODIS Land Science Team, 2020) shows classic CAOs on these days. In contrast, the other samples were associated with less well-defined air mass trajectories that were not CAO events or, in one case, with a CAO event where the air originated from the west in the Canadian archipelago rather than the central Arctic.

Geerts et al. (2022) reported INP concentrations derived using the wash-off technique during CAOs from December 2019 to May 2020 at Andenes on the coast of northern Norway. The ground-based measurements at this downstream site revealed INP concentrations that were 2–3 orders of magnitude lower than in ACAO. This might be consistent with loss of INPs in the precipitating clouds in the CAO. Ground measurements of INP concentrations made during 2012 by Wex et al. (2019) in Ny-Ålesund (upstream of the CAO clouds in ACAO), using a quartz filter technique, were also 2–3 orders of magnitude lower than those observed during ACAO. However, the highest INP concentrations observed in this dataset occurred during summer rather than springtime. A similar seasonal dependence was also observed in the high Arctic by Creamean et al. (2022), who observed INP concentrations 2–4 orders of magnitude below those in the ACAO campaign during their year-round INP measurements.

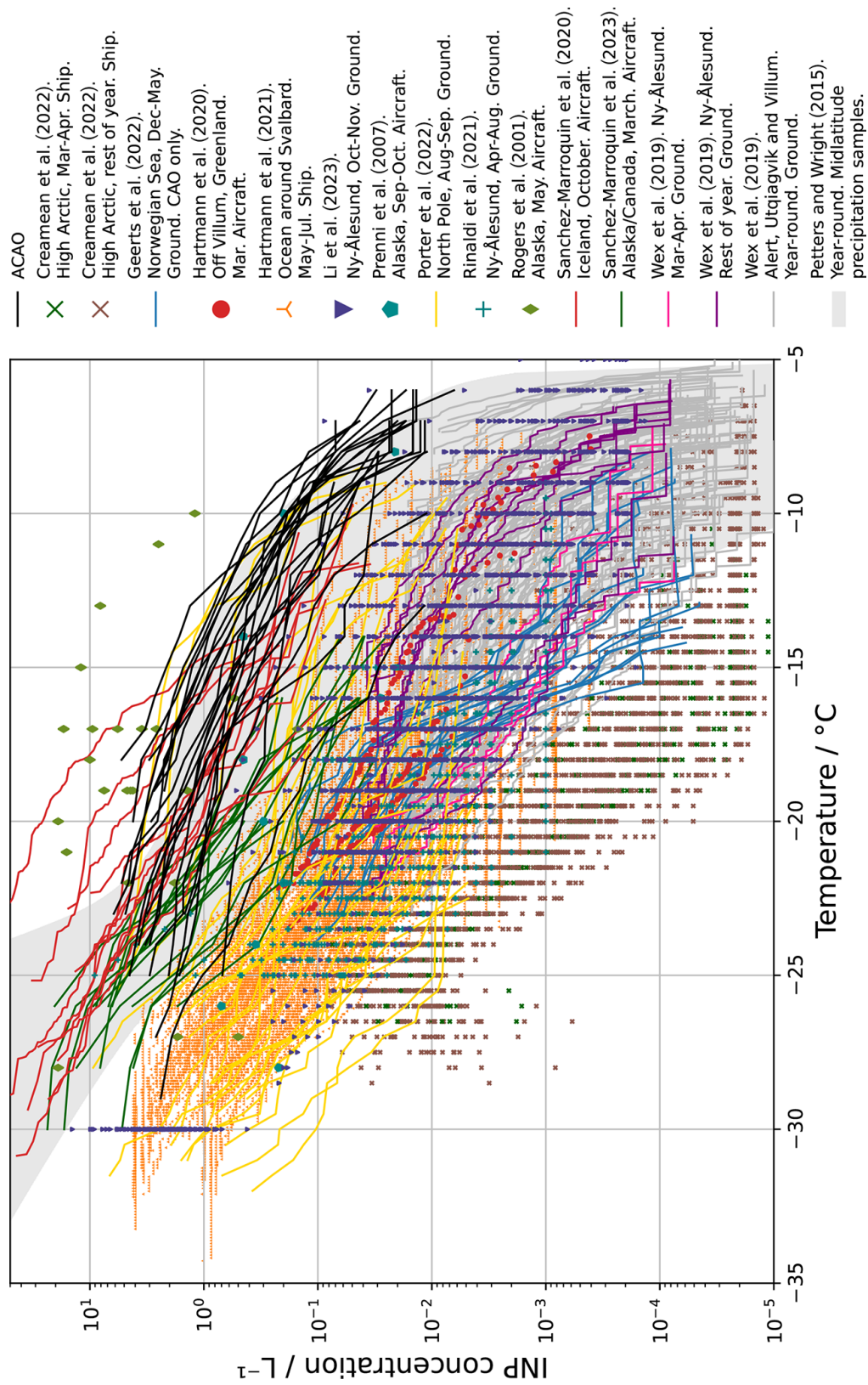


Figure 2.4: INP concentrations measured in a number of different field campaigns in the Arctic (Creamean et al., 2022; Geerts et al., 2022; Hartmann et al., 2020, 2021; Li et al., 2023; Prenni et al., 2007; Porter et al., 2022; Rogers et al., 2001; Rinaldi et al., 2021; Sanchez-Marroquin et al., 2020, 2023; Wex et al., 2019). These are compared to mid-latitude INP measurements from precipitation samples collated by Petters and Wright (2015).

Creamean et al. (2022) used a technique that involved size selecting aerosols, impacting them onto petrolatum and then recovering these particles into water for a droplet-freezing assay. Ship-based measurements of INPs in the ocean around Svalbard, made with the wash-off technique between May and July 2017, were 1–2 orders of magnitude below those made during the ACAO campaign (Hartmann et al., 2021).

Rinaldi et al. (2021) used two offline methods to measure INP concentrations sampled at a ground-based site near Ny-Ålesund between April and August 2018. They found that using a dynamic filter processing chamber to measure INP concentration using condensation freezing yielded INP concentrations approximately 8 times greater than those yielded by a droplet-freezing assay measurement using a wash-off technique. However, the highest concentrations measured by Rinaldi et al. (2021) were lower than the lowest observed during ACAO and were typically 1–3 orders of magnitude below those measured in this campaign. Similarly, Li et al. (2023) used several different techniques to measure INPs at a Ny-Ålesund ground site during October and November 2019. These included an impinger sampling method, a continuous-flow diffusion chamber (CFDC) and a polycarbonate filter wash-off technique. These measurements had a much greater spread than those in ACAO and concentrations were typically 1–4 orders of magnitude lower. However, the measurement spread reduced at lower temperatures, and if extrapolated log-linearly, wash-off measurements were consistent with measurements made by the CFDC at $T = -30\text{ }^{\circ}\text{C}$. Similarly, although the CFDC measurements were made at lower temperatures than the INP measurements in ACAO, the two were consistent if the ACAO spectra were extrapolated log-linearly as the gradient of INP spectra in ACAO was lower than that measured by Li et al. (2023).

Comparison with the INP literature presents a somewhat confusing picture. It may simply be the case that there is strong variability on a range of spatial and temporal scales. However, there may also be differences between techniques. As noted in the methods section, intercomparison of assay techniques reveals similar results for some INP types (DeMott et al., 2018; Wex et al., 2019) but substantial differences between different techniques for some INP types (Hiranuma et al., 2019). Additionally, intercomparison of assay techniques on simultaneously taken samples of ambient aerosol during field campaigns has shown both consistencies and inconsistencies between techniques (Li et al., 2023; Lacher et al., 2024). Beall et al. (2020) report that biological INPs can sometimes be lost during storage and transport, which we largely avoid by processing our filters during the campaign in a temporary laboratory. While the droplet-on-filter technique used here was not included in the formal intercomparisons, it has shown different responses in other environments, and the results are consistent with what might be expected. For example, this technique produced INP concentrations several orders of magnitude smaller than in ACAO during March 2018 near Alaska (Sanchez-Marroquin et al., 2023). In dust plumes emerging from Africa, this technique produced much steeper INP spectra than in ACAO (Price et al., 2018), and the INP concentrations were consistent with laboratory-derived activity of K-feldspar in mineral

dust (Harrison et al., 2019). Measurements in the UK revealed a strong biological “hump” consistent with ground-based measurements (Sanchez-Marroquin et al., 2021). In addition, measurements in dust plumes emerging from Iceland produced active site densities consistent with laboratory measurements of Icelandic dust (Sanchez-Marroquin et al., 2020) and other high-latitude dusts (Barr et al., 2023). Continued efforts are needed to intercompare INP assay techniques and better understand the influence of storage, but high Arctic INP concentrations have been observed by various groups using various techniques.

Overall, the literature in combination with our new measurements indicates that INP concentrations during northerly CAOs in this region, where the air is drawn from the central Arctic, can at least sometimes have mid-latitude-like INP concentrations.

2.3.2 Aerosol environment in CAOs

To try to understand the sources and variation in the high INP concentrations observed during the campaign, we used observations from the PCASP and CDP underwing probes to investigate the aerosol environments during filter sampling.

Size distributions of the aerosol particle number, N ; surface area, S ; and volume, V , are presented in Figure 2.5. To derive surface area and volume, aerosols were assumed to be spherical. The size distributions of aerosol measured during the campaign were mostly similar in shape but with values of $dN/d \log(D)$ that spanned 2 orders of magnitude. Number concentrations of aerosol were dominated by submicron particles. Few super-coarse aerosol particles (diameter $D > 10 \mu\text{m}$) were observed, though particles with diameters greater than $30 \mu\text{m}$ were at the edge of the detection ability of the CDP. Sub- and super-micron particles contributed approximately equally to the total surface area of the aerosol. Number concentrations of aerosol observed were consistent with those observed over the Norwegian Sea during the March–April 2013 ACCACIA aircraft campaign using the PCASP and CDP instruments by Young et al. (2016). However, the number concentration of sub-micron aerosol was up to 2 orders of magnitude greater than in March observations of aerosol made near Ny-Ålesund by Song et al. (2021) from 2015–2019 using a ground-based aerodynamic particle sizer. The number concentration of aerosol with a diameter between $0.1 \mu\text{m}$ and $0.5 \mu\text{m}$ observed during ACAO was also an order of magnitude greater than that measured at Ny-Ålesund during the COMBLE campaign, though concentrations were similar between $0.5 \mu\text{m}$ and $1 \mu\text{m}$ (Williams et al., 2024).

Figure 2.6 shows aerosol profiles from each of the flights where PCASP and CDP data were available. Measurements of aerosol concentration where filter samples were taken are highlighted according to their INP concentration, showing that the INP filter samples taken were subject to aerosol concentrations representative of the aerosol environment at these heights measured throughout the flight. Aerosol concentrations near the sea surface (less than 500 m above sea level) were consistently 100 cm^{-3} to 200 cm^{-3} . These measurements were comparable to measurements made in early April 2022 near Svalbard during the

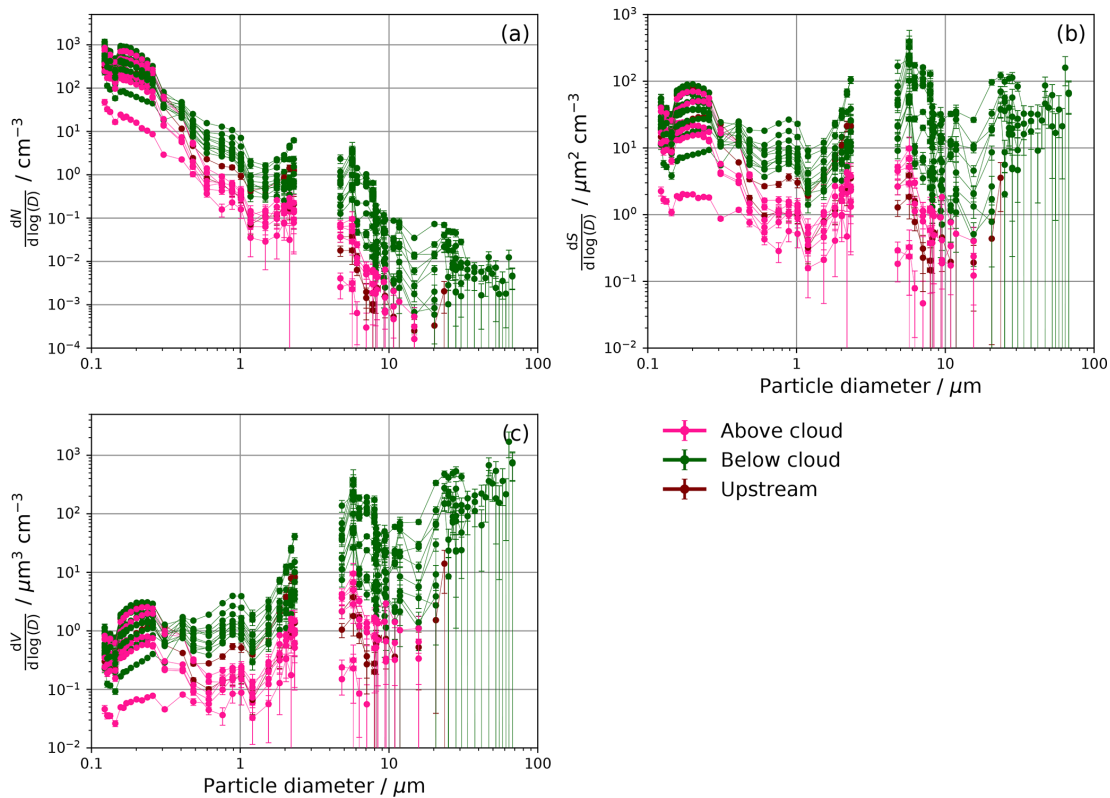


Figure 2.5: Size distributions of (a) particle number, (b) particle surface area and (c) particle volume measured with the PCASP and CDP probes during each filter sample. Probe data were unavailable during flights c271 and c272. A version of this figure where each size distribution is individually labelled according to the sample it is associated with can be found in Section A.2.

HALO-(AC)³ campaign (Wendisch et al., 2024). However, the vertical distribution of aerosol varied much more between flight days. On 3 flying days, aerosol concentrations were approximately constant from the surface up to 7500 m. These were c274, which had a northwesterly flow, and c277–c278 and c281–c282, which both had northerly flows. These contrasted with three days where the aerosol concentration decreased with altitude in the boundary layer. These were c273, which had a westerly flow, and c275–c276 and c279, which both had northerly flows. Finally, aerosol concentrations increased with altitude during flight c280, another day with a northerly flow.

2.3.3 Size-resolved aerosol composition

SEM analysis to determine size-resolved composition was performed on two polycarbonate filters sampled during filter runs c279r1 and c278r2. These filters were selected to represent the aerosol upstream of the CAOs and had an $N_{\text{INP}}(-15^\circ\text{C})$ of $(0.49 \pm 0.15) \text{ L}^{-1}$ and $(1.4 \pm 0.4) \text{ L}^{-1}$ respectively. These represent INP concentrations at the 33rd and 90th percentiles of samples taken in CAOs. Filter c279r1 was sampled at an altitude of 155 m over the marginal sea ice zone, while filter c278r2 was sampled at an altitude of 1725 m above the Svalbard archipelago and surrounding sea ice, prior to cloud development. The size

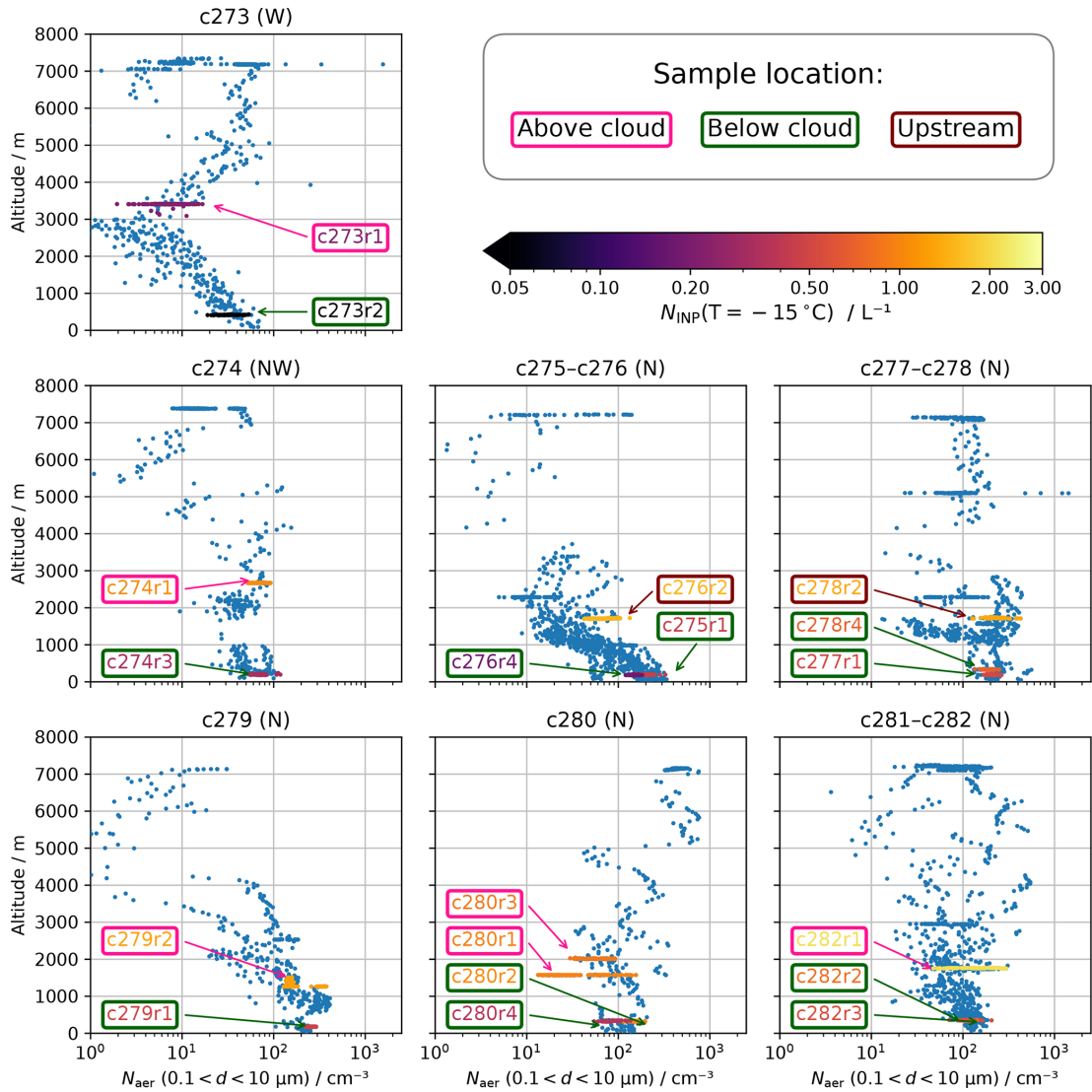


Figure 2.6: Out-of-cloud aerosol measurements made using the PCASP and CDP averaged every 10 s. Measurements made during filter sampling are coloured and labelled according to their INP concentration at $T = -15^\circ\text{C}$. The borders of each label and the arrows matching labels to measurements are coloured according to the location of the sample relative to the CAO cloud. Measurements over or close to the Scandinavian Peninsula were discarded to represent Arctic and oceanic air only.

and elemental proportions of 1101 and 451 particles respectively were analysed on these filters.

Composition analysis and particle size distributions derived using SEM for each of the filters are shown in Figure 2.7. Figure 2.7 also compares the particle size distributions derived using SEM with those derived from the PCASP and CDP. Both filters showed that a large proportion of the particles were consistent with mineral dusts, with Si-rich and Al- and Si-rich particles representing over half of the proportion of supermicron particles. There were also significant numbers of carbonaceous aerosol in the submicron range, although this method cannot confirm if they were of biological origin. We did not note any particles that were qualitatively of biological origin on the basis of size or morphology, as noted previously in Sanchez-Marroquin et al. (2021). However, due to the relatively small fraction of filter analysed, their presence cannot be ruled out. Despite filter sample c279r1 being taken close to the marginal ice surface, less than a quarter of the particles showed clear signatures of sea spray (such as the presence of both sodium and chlorine), though some of the carbonaceous or sulfurous aerosol could be of marine origin. However, the sea ice coverage is likely to have inhibited the generation of sea spray. The total surface area of mineral dust particles, being the sum of Si-rich, Si- and Al-rich, Si-only, and Ca-rich classes, was calculated as $1.0 \mu\text{m}^2 \text{cm}^{-3}$ and $0.6 \mu\text{m}^2 \text{cm}^{-3}$ for c279r1 and c278r2 respectively. These values are consistent with measurements of aerosols 91 m to 375 m over the Norwegian Sea in March, made by Young et al. (2016) during the ACCACIA campaign. Filter c279r1 showed very few particles with clear sea-spray signatures, consistent with its higher altitude and location over sea ice. Metal-rich particles are present in the smaller-size fraction, although these have previously been found to be contaminant particles on blank filters. These were likely produced by the microscope mechanism, so it is not clear if they are artefacts or from the atmosphere.

The aerosol number measured by the SEM is generally 2–4 times below the aerosol number reported by the PCASP and CDP probes or shifted to about 30 % smaller sizes. Previous studies using this technique have also found similar magnitudes of discrepancy between the techniques. In these cases, SEM has sometimes measured more particles than the optical probes and sometimes less (Sanchez-Marroquin et al., 2020, 2021). Using a different SEM technique, Young et al. (2016) found the PCASP and CDP probes reported concentrations up to 100 times greater than SEM and attributed this larger discrepancy to measurements of cloud droplets or swollen aerosol since they sampled at high RH (Φ), often with $\Phi > 90\%$. We discarded PCASP measurements made in cloud or above $\Phi = 80\%$ to bias against swollen aerosol. However, even at $\Phi = 80\%$, the diameter growth factor of typical organic aerosol has been reported to be between 20–50 % (Martin et al., 2003; Latimer and Martin, 2019). For sea salt, the diameter growth factor at $\Phi = 80\%$ has been reported to be between 60–100 % (Tang et al., 1997; Martin et al., 2003; Murray et al., 2012a). Hygroscopic growth of these species might help explain our discrepancy. However, the measured size and number of the mineral dust particles should not be affected by humidity.

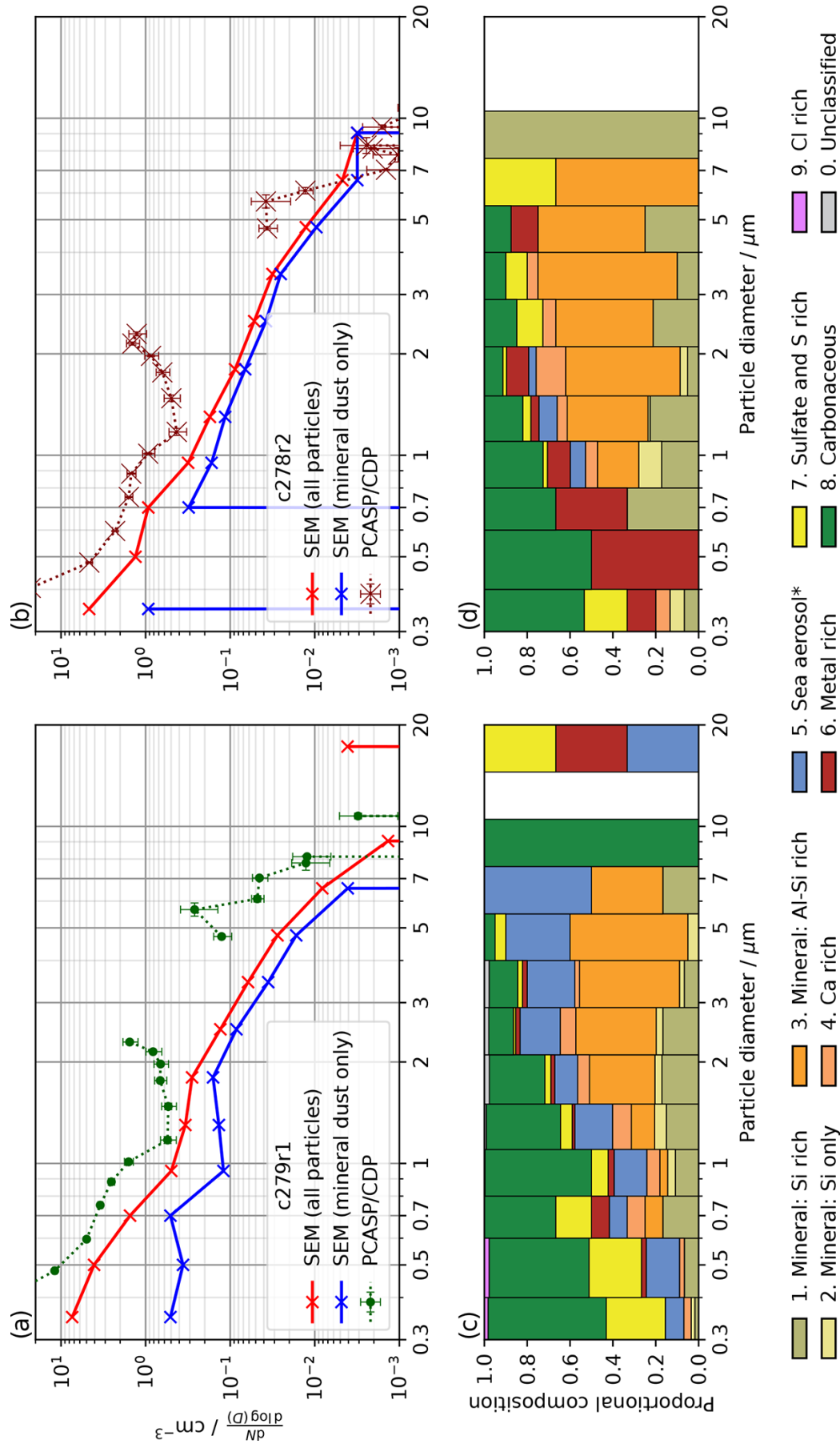


Figure 2.7: Particle size distributions and particle composition derived from scanning electron microscopy for filters c279r1 (a, c) and c278r2 (b, d). Sample c278r2 was taken upstream of cloud development, while sample c279r1 was taken below the northern extent of CAO cloud streets. Both were taken in northerly CAOs. The solid lines show particle size distributions derived directly from the SEM measurements, while the dotted lines show the particle size distribution derived from the underlying probes in the diameter ranges, as in Figure 2.5. Composition is shown for each size bin. Size bins with greater values of $dN/d \log(D)$ contain more particles. The white regions of the composition plots indicate that no particles were detected in this size range. Sea aerosol refers to aerosols likely to correspond to fresh, aged or mixed sea-spray aerosols.

2.3.4 Ice-nucleating activity of samples

To determine how ice-nucleating activity depends on the physical properties of the aerosols, the aerosol size distributions presented in Figure 2.5 were used to calculate the total aerosol number N_{aer} , surface area S_{aer} and volume V_{aer} for aerosols with $D > 0.1 \mu\text{m}$, assuming spherical geometry. Figure 2.3 shows the INP concentrations normalised by aerosol number, area and volume. Normalising N_{INP} by N_{aer} failed to reduce the spread in values between measurements, while normalising by S_{aer} and V_{aer} increased the spread between measurements. This indicates that the INP concentration does not scale with the overall aerosol surface area or volume. This is consistent with the INPs being a subset of the aerosol particle population, with non-ice-nucleating components of the aerosol particle population being the primary driver of variability in S_{aer} and V_{aer} . Other studies of Arctic and high-latitude INPs (e.g. Porter et al., 2022; Barr et al., 2023; Sanchez-Marroquin et al., 2023; Moore et al., 2024) have also found that normalising by surface area fails to reduce the measurement spread.

In Figure 2.8, measurements of $N_{\text{INP}}/S_{\text{aer}}$ are compared to measurements of active site density reported for well-defined samples and from other field campaigns. Both the magnitude and the shape of the curve of $N_{\text{INP}}/S_{\text{aer}}$ values from ACAO are similar to those measured by Porter et al. (2022) at the North Pole in late summer. This indicates that there is perhaps some commonality in INPs between these two campaigns, even though the timing and location were very different.

To compare $N_{\text{INP}}/S_{\text{aer}}$ to the active site density for well-defined samples, such as mineral dusts or sea spray, we note that these comprise only a fraction of the surface area in the aerosol samples in ACAO (see Figure 2.7). In the case of sea spray, our $N_{\text{INP}}/S_{\text{aer}}$ values are many orders of magnitude greater than the active site density for pristine sea spray reported by McCluskey et al. (2018b); hence we conclude that the INPs we observe are not related to pristine sea spray.

Integration of the size distributions in Figure 2.7 reveals that 32 % (c279p1) and 53 % (c278p2) of the overall aerosol surface area was mineral dust. Assuming that the aerosol composition was similar across the campaign, values of $N_{\text{INP}}/S_{\text{aer}}$ would be 2–3 times smaller than values of active site density normalised to the mineral dust surface area rather than the overall aerosol surface area. This allows us to compare with literature parameterisations of active site density for K-feldspar, desert dusts and soil dusts containing ice-active organic material in Figure 2.8. Comparison with K-feldspar, the most active mineral component of atmospheric mineral dusts (Harrison et al., 2019; Vergara-Temprado et al., 2017), shows that K-feldspar alone cannot account for the magnitude of the observed active site densities above -20°C as the temperature dependence is too steep. This implies that there is some other component in these samples that nucleates ice. Above -20°C , measurements fall between the active site densities reported for agricultural soil by Steinke et al. (2016), fertile soils by O’Sullivan et al. (2018) and general mineral dusts (from fertile and infertile

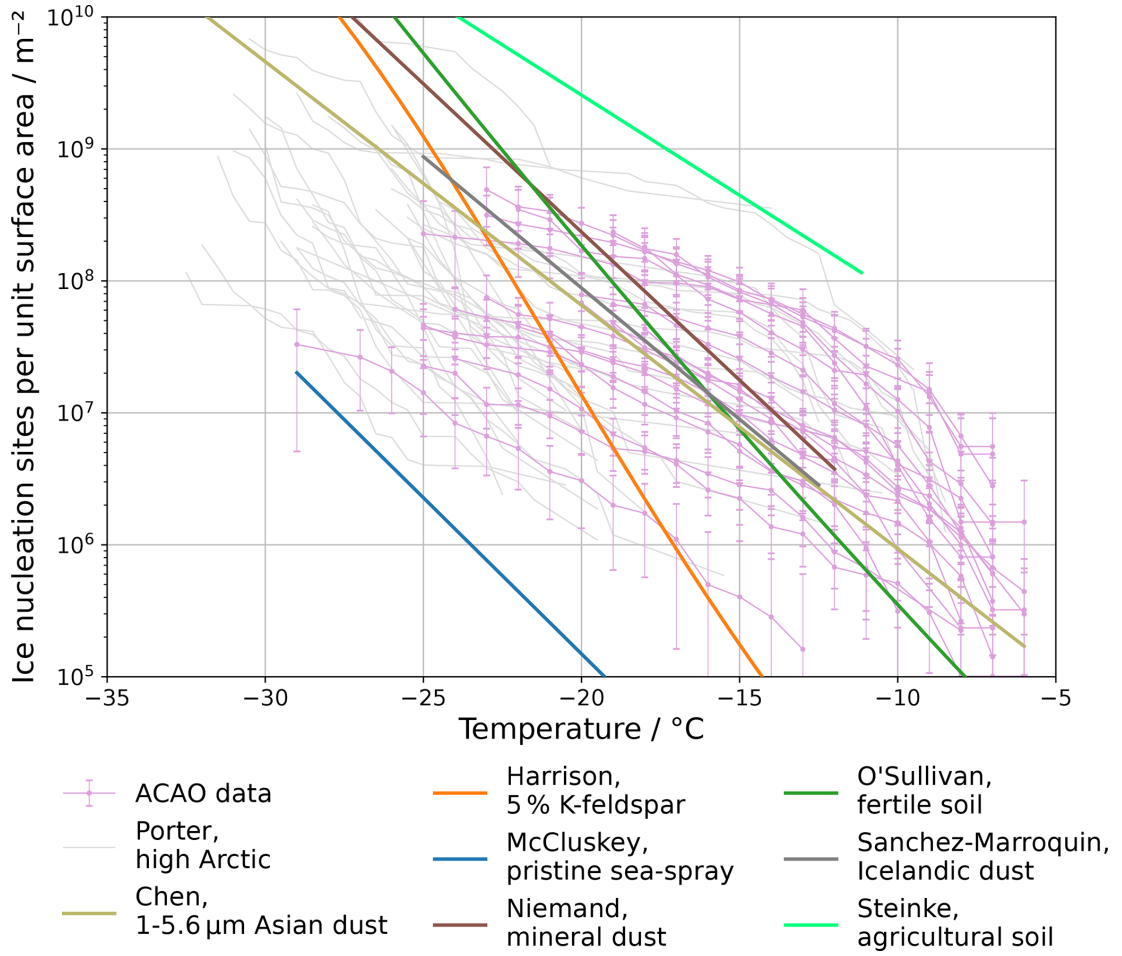


Figure 2.8: A comparison of $N_{\text{INP}}/S_{\text{aer}}$ measured during the ACAO campaign to measurements of active ice nucleation site density in the high Arctic by Porter et al. (2022) and parameterisations of active ice nucleation site density (Chen et al., 2021; Harrison et al., 2019; McCluskey et al., 2018b; Niemand et al., 2012; O'Sullivan et al., 2018; Sanchez-Marroquin et al., 2020; Steinke et al., 2016).

sources) by Niemand et al. (2012). The measurements are also similar to the active site densities for supermicron Asian dust reported by Chen et al. (2021) and Icelandic dust measured by Sanchez-Marroquin et al. (2020).

In conclusion, the high values of $N_{\text{INP}}/S_{\text{aer}}$ of the ACAO samples at temperatures greater than -20°C suggest a strong biogenic component to the INP population (Huang et al., 2021), potentially corresponding to mineral dust containing biological components, such as fertile soils.

2.3.5 Variation in INP properties with altitude and position in CAOs

To understand how the INP spectra varied with altitude during ACAO, we considered the variation of INP properties with altitude in northerly CAOs (flights c274 to c282). In Figure 2.9, $N_{\text{INP}}(-15^{\circ}\text{C})$, the INP concentrations at -15°C , as well as the same quantity normalised by surface area, are presented relative to the altitude of the samples. This temperature was

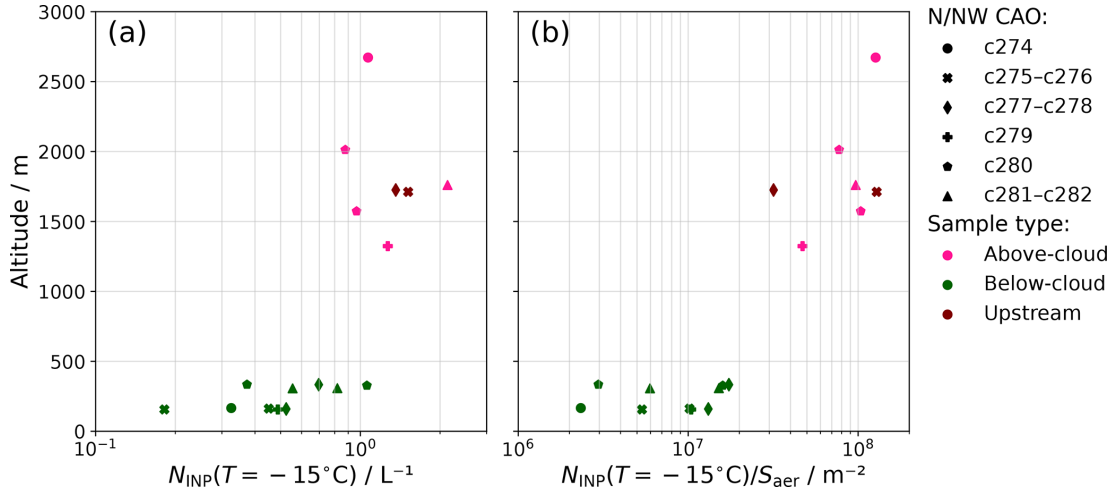


Figure 2.9: Variation with altitude of (a) INP concentration at -15°C and (b) INP concentration at -15°C normalised by aerosol surface area in northerly CAOs.

Flight	Upwind filter	Downwind filter	$\Delta\phi$	F_{INP}	F_{S}	F_{aer}
c275-276	c275r1, Sc	c276r4t, Sc	1.92	0.40	0.52	0.75
c280	c280r2, Sc	c280r4t, precipitating Cu	2.17	0.35	0.19	1.85
c281-282	c282r2, Sc	c282r3t, precipitating Sc-Cu	3.58	0.68	0.39	1.69

Table 2.2: Factors of change in the INP concentration at -15°C (F_{INP}), INP concentration normalised by surface area at -15°C (F_{S}) and total aerosol surface area (F_{aer}) between three pairs of below-cloud filter measurements.

chosen to be representative of typical cloud temperatures in ACAO and because spectra with relatively high INP concentrations at -15°C had similarly high concentrations relative to other INP spectra in the campaign across all temperatures.

Considering $N_{\text{INP}}(-15^{\circ}\text{C})$, the samples fall into two groups, as shown in Figure 2.3. Samples taken above the clouds and upstream of the clouds fall into a group where the mean value of $N_{\text{INP}}(-15^{\circ}\text{C})$ was 1.3 L^{-1} above the cloud, whereas the below-cloud value was 0.54 L^{-1} . This indicates that the below-cloud INP concentrations are approximately 2.4 times smaller than the above-cloud measurements. This is consistent with a free tropospheric source of INPs rather than a source from the surface. It is also possible that there was a removal of INPs by clouds, which we return to below. When normalised to the aerosol surface area, the gap between the two groups is even larger, with above-cloud values of $N_{\text{INP}}(-15^{\circ}\text{C})/S_{\text{aer}}$ approximately 8.8 times larger than the below-cloud values. Similar relationships between INP activity and altitude were found by Moore et al. (2024), who observed that INP concentrations normalised by surface area above the cloud in the SOCRATES campaign were greater than those below the cloud. Additionally, Knopf et al. (2023) found that during measurements on INPs in the eastern North Atlantic, the efficiency of deposition ice nucleation was greater for samples taken in the free troposphere than in the marine boundary layer. The reduced $N_{\text{INP}}(-15^{\circ}\text{C})/S_{\text{aer}}$ below the cloud is due to the enhanced aerosol surface area below the cloud (Figure 2.5) in the CAO that was most likely related to enhanced sea-spray

production. White caps on waves were visible downstream in the CAO throughout the campaign and mean 10 m wind-speeds over the sea in the sea ice free region ranging between 8 m s^{-1} and 12 m s^{-1} , while maximum 10 m wind-speeds were between 15 m s^{-1} and 25 m s^{-1} (C3S, 2018). The size distribution of below-cloud aerosol is of similar scale to those produced by the sea-salt aerosol source parametrisation of Gong (2003), suggesting that aerosol number was dominated by sea spray downstream. The reduction in ice-nucleating activity is consistent with this sea spray producing a negligible concentration of INPs (see Section 2.3.4) and so contributing additional aerosol surface area with limited ice-nucleating ability. Using the average below-cloud aerosol surface area with the parameterisation of McCluskey et al. (2018b), the INP concentration at -15°C would be expected to be $4 \times 10^{-4} \text{ L}^{-1}$ if the only INPs below the cloud were sea spray. Our measured INP concentration is more than 3 orders of magnitude greater than this, again indicating that sea spray is not an important INP type in these CAO events.

On three flights (Table 2.2), two below-cloud filter samples were taken at different points in the CAO. We might expect aerosol and INP scavenging to increase further south as CAOs develop, especially in the cumulus regime where intense precipitation might efficiently remove particles. A downwind decrease in aerosol number concentration in Norwegian Sea CAOs attributed to precipitation scavenging has previously been observed by Williams et al. (2024). Using the values of $N_{\text{INP}}(-15^\circ\text{C})$ and the total aerosol surface area S_{aer} , we calculated the factors of change in INP concentration (F_{INP}), ice-nucleating activity (F_{S}) and surface area (F_{aer}) between the upwind and downwind measurements:

$$F_{\text{INP}} = \frac{N_{\text{INP}}(-15^\circ\text{C}, \text{downwind})}{N_{\text{INP}}(-15^\circ\text{C}, \text{upwind})}, \quad (2.5)$$

$$F_{\text{S}} = \frac{N_{\text{INP}}(-15^\circ\text{C}, \text{downwind})/S_{\text{aer}}(\text{downwind})}{N_{\text{INP}}(-15^\circ\text{C}, \text{upwind})/S_{\text{aer}}(\text{upwind})}, \quad (2.6)$$

$$F_{\text{aer}} = \frac{S_{\text{aer}}(\text{downwind})}{S_{\text{aer}}(\text{upwind})}. \quad (2.7)$$

These values are displayed in Table 2.2 with the difference in latitude between the measurements, $\Delta\phi$. Both the INP concentration and the ice-nucleating activity decreased with latitude. The total surface area of the aerosol increased in two flights by 69 % and 85 % but decreased by 25 % in one pair. The upwind filters were sampled under stratocumulus conditions, while the downwind filters were in a mixture of stratocumulus and cumulus, with precipitation in two cases (c280r4 and c282r3).

During flight c280, two pairs of above- and below-cloud filter samples were taken along the same wind trajectory (Figure 2.1b). The northern pair was taken where the clouds had stratocumulus forms, whereas the southern pair was taken where the clouds were in the cumulus regime. Comparing the INP spectra for these samples in Figure 2.10 shows that except at the very highest temperatures, where the error is large, there is little difference between the samples taken above and below the stratocumulus (c280r1 and c280r2 respect-

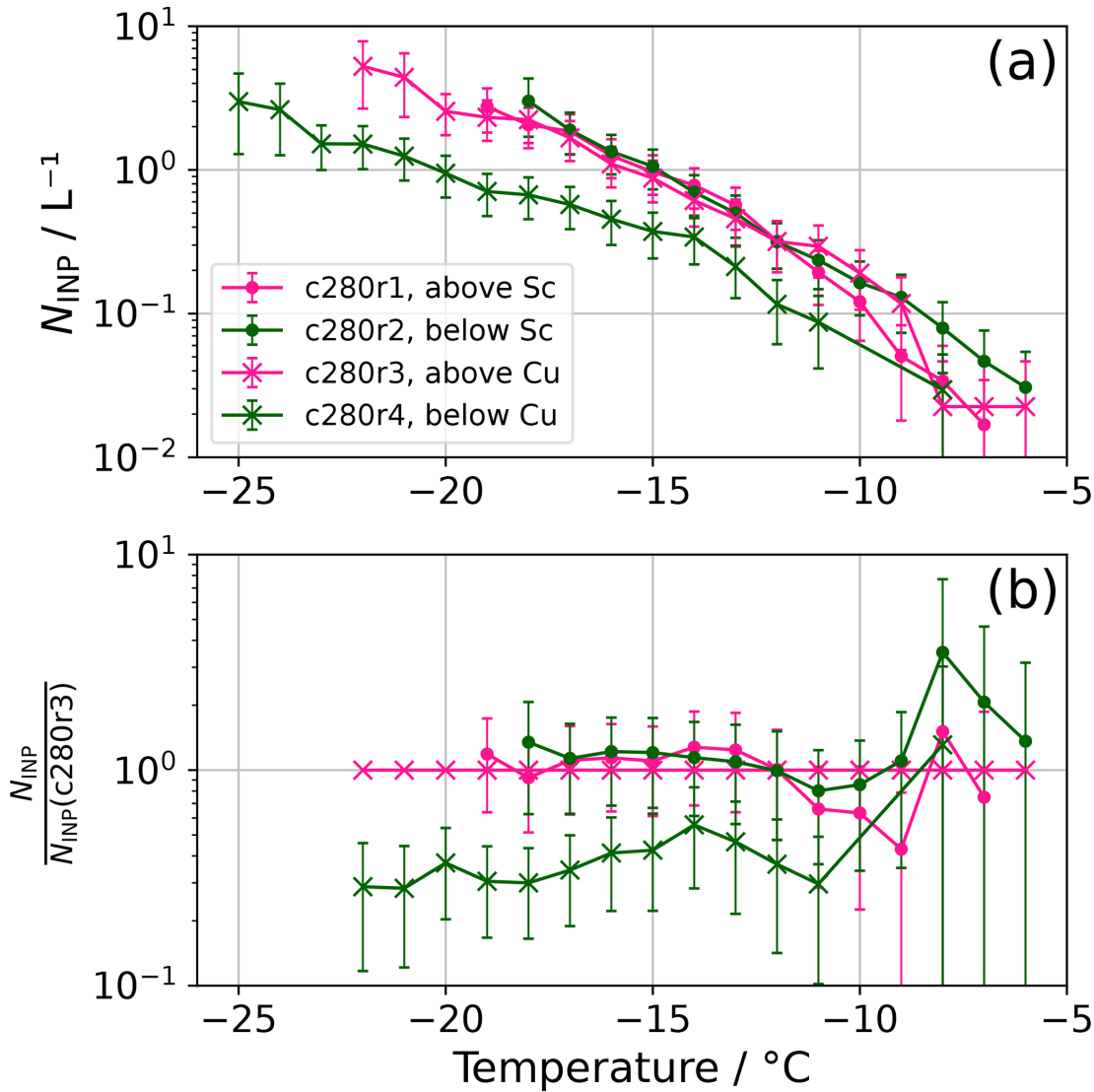


Figure 2.10: Panel (a) shows the four filter samples taken during flight c280 and describes their location relative to either stratocumulus (Sc) or cumulus (Cu) clouds. Panel (b) shows their INP concentrations relative to c280r3 (the sample taken above the cumulus) for the temperature ranges over which they can be compared.

ively) and the sample taken above the cumulus (c280r3). Closeness between the three INP measurements may be expected if the main INP source entering the system is upwind of the CAO clouds. However, the southern sample taken below the cumulus (c280r4) has over 5 times fewer INPs at temperatures below -11 °C.

Together, these results are consistent with INPs being removed by cloud and precipitation scavenging in the CAO, although this conclusion is based on only three cases. If precipitation scavenging is strong, it is possible that the INP concentrations measured during ACAO could be consistent with the INP concentrations measured at Andenes by Geerts et al. (2022). As such, it would be helpful to test this with future aircraft INP measurements much deeper into the cumulus regime (i.e. further south). This was not feasible in ACAO due to a com-

bination of unfavourable meteorology, airspace restrictions and the prioritisation of tasks within the available flight time.

2.3.6 Backward-trajectory analysis

To investigate the potential sources of sampled aerosol, backward trajectories were produced for each sample using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015). Meteorological data from the Global Data Assimilation System (GDAS) at 1° resolution were used to obtain the trajectories. The backward trajectories were run for 7 d and initiated along each flight track at 3 min intervals during each of the sampling periods to produce multiple trajectories. Figure 2.11a shows these trajectories, and Figure 2.11b shows the INP concentration at -15°C associated with each air mass. The majority of the air masses associated with northerly cold-air outbreaks circulated in the high Arctic for several days. Air masses associated with westerly CAOs (c273, c274) typically passed over Greenland, the Atlantic Ocean or Canada. Some of the air masses associated with northerly CAOs passed over Asiatic mid-latitudes 6 d to 7 d prior to sampling. However, there was no clear correlation between INP concentration and air mass origin, suggesting that the air masses did not interact with a distinct source of INPs in 7 d transport. This might be consistent with a diffuse INP source across the whole of the Arctic region or INPs that were emitted further back in time than 7 d ago and transported into the Arctic where they circulated around in the Arctic's stable atmosphere prior to being drawn out in CAO events. We return to these ideas in the next section.

2.4 Discussion of potential INP sources

The INP concentrations measured during the ACAO campaign were amongst some of the highest measured in the Arctic (Figure 2.3 and associated discussion). We now discuss several possible explanations for these high concentrations and thus the potential sources of INPs.

If the INPs were of local origin, strong local sources of INPs would be required to explain the high INP concentrations. Potential local sources of INPs in the region of the Norwegian Sea (Figure 2.1a) include upstream INP sources from Arctic land such as the Svalbard archipelago; continental INPs from Europe; and sea-spray aerosol from the open sea, polynyas and open leads. However, there are several reasons why these Arctic sources are unlikely to play a major role.

The presence of dust indicated by the SEM analysis and the consistency of the measured active site densities shown in Figure 2.8 with those previously measured for biologically enhanced mineral dusts could indicate a local terrestrial source of INPs. It has been shown that mineral dusts from high-latitude sources contain ice-active biogenic materials (Barr et al., 2023; Tobo et al., 2019; Xi et al., 2022); hence dust might be sourced from the Sval-

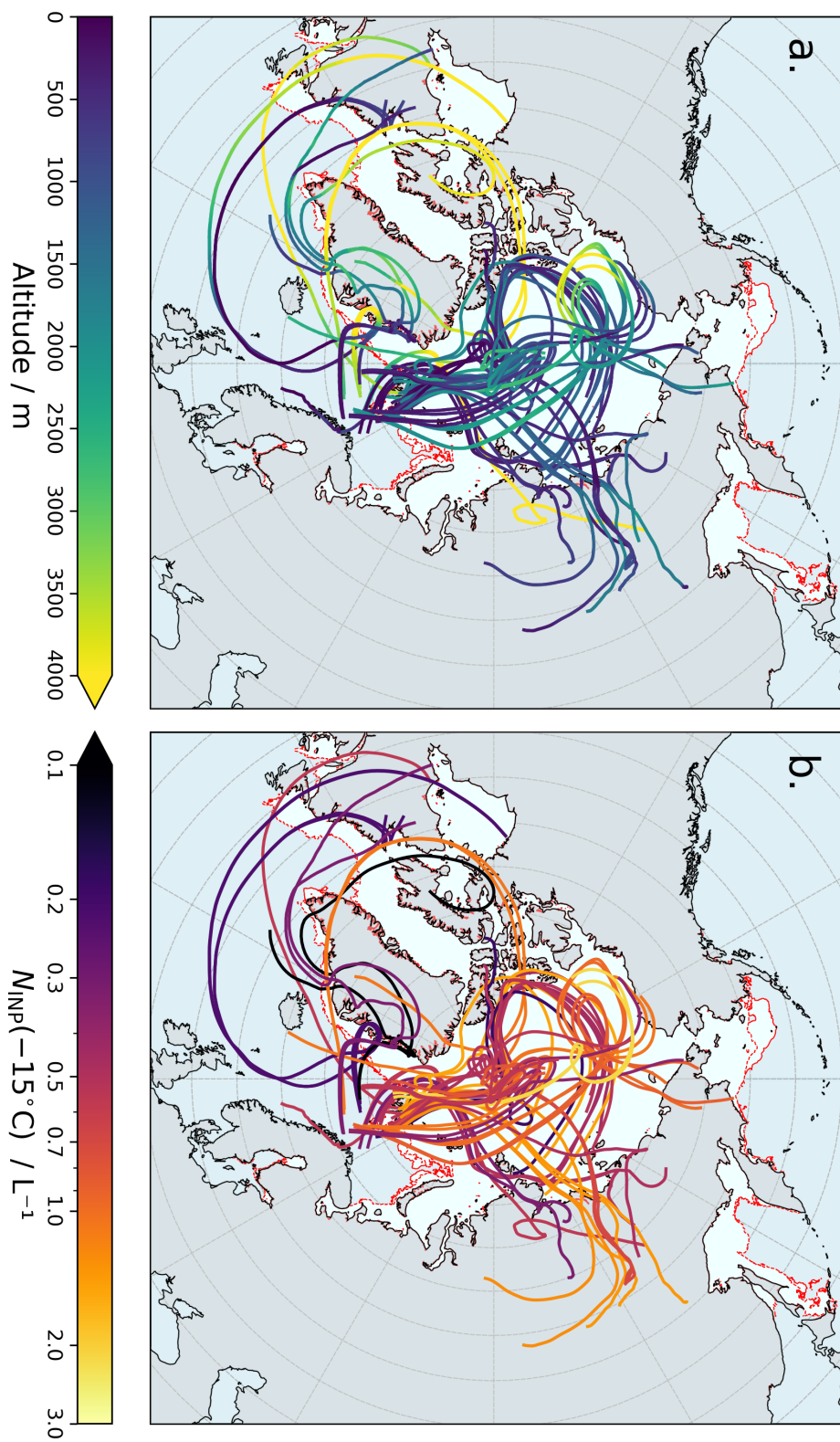


Figure 2.11: The 7 d backward trajectories of air masses sampled. Two trajectories are shown for each sample, corresponding to the start and end of the sampling periods. There was little variation in trajectories at intervals within the sampling periods. Panel (a) shows the altitudes associated with the trajectories, while panel (b) shows the relative magnitude of INP concentration. Backward trajectories from c272r6, measured in a warm-air intrusion, are not included.

bard archipelago or Greenland. However, back-trajectory analysis does not indicate that air masses directly from Svalbard or Greenland are consistently associated with the most active INP samples. Additionally, the pristine snow cover over land in March means that emissions of aerosol are strongly suppressed and are therefore unlikely to dominate the INP population observed during ACAO (Bullard et al., 2016; Meinander et al., 2022). Although there have been reports of dust emission events in Svalbard happening as early in the year as May (Dörnbrack et al., 2010), these events in Svalbard are typically reported in autumn when snow cover is at its minimum (Meinander et al., 2022; Rymer et al., 2022).

It is also unlikely that transport of mid-latitude continental INPs from Europe directly into the CAO regimes was a dominant source of INPs since the CAO conditions exhibited northerly or westerly flows. Early in the campaign, prolonged warm-air intrusion conditions exhibited southwesterly winds. However, some of the INP concentrations measured in the cold-air outbreak immediately after the warm-air intrusion had subsided (c273r2) were the lowest measured in the campaign.

Several aspects of the measurements suggest that sea-spray aerosol from the open sea was neither the dominant INP source nor a significant contributor to the ice-nucleating activity. Neither of the aerosol samples analysed with SEM showed that aerosols with clear sea-spray origins dominated the aerosol number, with less than 20 % of the aerosol in the sample taken at an altitude of 155 m over the marginal ice zone having a sea-spray origin. In the boundary layer, the measured INP concentrations were several orders of magnitude higher than those measured in similarly remote areas in the Southern Ocean or the North Atlantic where sea-spray aerosol is the dominant INP source (McCluskey et al., 2018a,b; Schmale et al., 2019; McFarquhar et al., 2021; Tatzelt et al., 2022). Furthermore, while the INP concentrations were similar above and below the cloud decks, the samples with the highest active site density were found above the cloud systems and in particular upstream of the outbreak (Figure 2.9), environments unlikely to have large sea-spray components. Considering all these aspects, it is unlikely that sea-spray aerosol was a dominant source of INPs.

A final potential local source of INPs is the sea ice and leads in the sea ice. It has previously been hypothesised that open leads could be a significant source of INPs in the Arctic (Bigg and Leck, 2001). However, observed INP concentrations from ship campaigns passing through leads have found INP concentrations several orders of magnitude below those measured in ACAO (Bigg and Leck, 2001; Creamean et al., 2022). Furthermore, backward trajectories from the high Arctic measurements of INPs by Porter et al. (2022) showed that air masses which had spent the majority of the past week over sea ice had the lowest INP concentrations. Snow blown from the surface of sea ice has been shown to be a source of sea salt aerosol (Yang et al., 2008; Gong et al., 2023), though there has been little investigation into this mechanism as a potential INP source, and the reduced activity of INP samples below the cloud deck again suggests that this would be unlikely to dominate the INP population. Together, these suggest that sea ice was not a dominant source of INPs during the

ACAO campaign.

Further evidence against a local surface source of INPs is that INP concentrations in northerly CAOs were fairly constant with altitude up to 3 km (Figure 2.9). A local surface source would lead to enhanced INPs at the surface in the highly stable stratified atmosphere of the Arctic in winter and early spring. However, high INP concentrations are persistent during all flights between 21 and 30 March across a wide area. Additionally, there are no large-scale ascending air masses exiting the Arctic that would carry the aerosol to 3 km altitude, and convection is limited to altitudes lower than 2.5 km even in well-developed CAOs.

Since it is unlikely that the dominant INPs had a local terrestrial or marine source, it follows that the high INP concentrations are likely to have originated from long-range transport. During the boreal winter, aerosol in the Arctic atmosphere can then have lifetimes of the order of weeks or months as a result of the dry and stable conditions leading to weak aerosol scavenging (Carslaw, 2022b). Coupled to this, we know that transport into the Arctic over the winter months leads to the build up of pollution, predominantly from Eurasian sources, which gives rise to the phenomenon of Arctic haze (Shaw, 1995; Stohl, 2006; Ekman and Schmale, 2022). While the anthropogenic component of Arctic haze has received substantial attention, it is also well known that other mid- and low-latitude aerosols are transported into the Arctic along with this pollution aerosol, such as mineral dust from African and Asian deserts (Rahn et al., 1977; Shi et al., 2022). Barrie (1986) suggested that the Arctic front extends as far south as 40°N in winter over the Eurasian landmass, encompassing sources such as Asian deserts and numerous pollution sources. Arctic haze layers have been reported to occur up to approximately 9 km altitude and have spacial extents of the order of 1000 km (Barrie, 1986). Our measurements of a widespread and vertically uniform distribution of INPs exiting the Arctic during the second half of March 2022 are consistent with a vertically extensive “reservoir” of INPs in the Arctic that is similar to pollution-related haze.

Previous airborne measurements of Arctic haze by Borys (1989) have suggested that Arctic haze, when identified by large signatures of anthropogenic pollutants, had 10 to 100 times fewer active INPs at -25 °C than Arctic aerosol samples with fewer pollutants. However, these low-pollutant samples may have been transported into the Arctic from low-latitude INP-rich environments, like desert regions, without being mixed with anthropogenic sources. Accordingly, aerosol traditionally defined as Arctic haze due to its large pollution signature may not be associated with aerosol of lower-latitude origins that has a high INP content. Springtime measurements of dust over the Norwegian Sea by Young et al. (2016) during the ACCACIA field campaign were attributed to long-range transport and had elemental composition ratios similar to those from Asian dust. During the ACCACIA campaign, plumes of aerosol from eastern and northern Asia were observed, which were hypothesised to have transported dust to the region (Liu et al., 2015; Young et al., 2016). As mentioned above, Asian dust has been thought to be a source of Arctic haze since the 1970s (Rahn et al., 1977), while modelling by Shi et al. (2022) has shown that during the

spring months, less than 20 % of the Arctic dust load above 900 hPa was emitted into the Arctic and that approximately 50 % of the dust load at all heights came from Asia. Modelling by Groot Zwaaftink et al. (2016) also attributed approximately 38 % of the Arctic dust load to Asian sources. This is significant since Asian dust is known to contain ice-active biological components (Creamean et al., 2013; Chen et al., 2021), which might be consistent with high ice-nucleating activities akin to those observed in ACAO. Transport routes for such biogenic-containing Asian dust have been demonstrated by Zhao et al. (2022), while year-round lidar observations by Ansmann et al. (2023) in the high Arctic have suggested that a small fraction of dust aerosols may control ice nucleation outside of summer. Based on the above evidence, we suggest that the dominant INP observed in ACAO was mineral dust internally mixed with ice-nucleating biogenic materials that originated from the lower latitudes and was transported into the Arctic through similar transport routes to Arctic haze.

The hypothesis of an INP reservoir appears to contradict previous remote sensing studies that used observations of Arctic clouds to infer spatial variations in INP concentrations. For instance, Carlsen and David (2022) define a temperature, T^* , at which mixed-phase clouds become more frequent than liquid clouds and use this as a proxy for freezing initiated by INPs. They find that in winter, when Arctic haze builds up, T^* is lower over the sea ice than over the open sea, which they attributed to a suppression of INP emissions (and concentrations) caused by sea ice cover. However, differences in ice concentrations between ice-covered and ice-free regions in high Arctic clouds may not reflect differences in INP concentration but rather differences in cloud microphysics related to cloud dynamics. For example, clouds over ice are shallow, long-lived and relatively stable compared to the deeper clouds with colder tops over ocean regions (Morrison et al., 2012; Arteaga et al., 2024). The long lifetime of these Arctic clouds provides time for INPs to be scavenged from the atmosphere; thus their small ice content may be a consequence of their history rather than the INP concentration when they first formed. Similarly, Murray-Watson and Gryspeerdt (2024) used satellite observations to suggest that increasing ice concentrations downwind in CAOs imply increased INP concentrations downwind, associating this with increased emissions of sea-spray INPs. This contrasts with our linked in-situ measurements of below-cloud INPs that reveal a decrease in INP concentration downwind (Table 2.2). The increased downwind ice concentrations observed by Murray-Watson and Gryspeerdt (2024) may be related to secondary ice processes, rather than INPs, since these are associated with strongly convective clouds, which are more likely to occur downwind. Additionally, clouds later in the CAOs tend to have lower cloud-top temperatures, naturally increasing the concentrations of ice.

2.5 Summary and conclusions

We made the first airborne measurements of INPs targeted at cold-air outbreaks over the Norwegian and Barents seas. We took 23 filter measurements and performed droplet-

freezing assays using a droplet-on-filter technique to determine INP concentration. INP concentrations measured over a large area of the Norwegian and Barents seas at altitudes between 40 m and 3400 m were amongst the highest reported in the Arctic, with values of $N_{\text{INP}}(-15\text{ }^{\circ}\text{C})$ ranging from 0.18 L^{-1} to 2.1 L^{-1} when the airflow had a strong northerly component. This indicates that high INP concentrations were horizontally and vertically widespread in these northerly flows, conducive to CAOs and boundary layer cloud formation, and persisted for at least 12 d in late March. These high INP concentrations contrast with previous surface measurements made in the same region and at the same time of the year but overlap with other airborne measurements using an online measurement technique. The lowest INP concentrations we measured during ACAO, $N_{\text{INP}}(-15\text{ }^{\circ}\text{C}) = 0.03\text{ L}^{-1}$, were in a westerly CAO. These results, combined with the literature data, highlight the importance of targeting INP measurements during specific meteorological events since an average INP concentration for this region and time would probably not represent the INP population for specific CAO clouds well.

To investigate the source of the high INP concentrations, we used particle size data from the underwing probes of the aircraft to normalise the INP concentration by the total aerosol surface area to gain an estimate of the density of active ice nucleation sites across the samples. Comparing this to literature measurements of active site density suggested that the INP population was likely dominated by mineral dust with ice-active biogenic components. Additionally, the samples with the highest ice-active site densities were found above the cloud system, suggesting that the INP population was not dominated by sea-spray aerosol. Scanning electron microscopy with energy-dispersive X-ray spectroscopy on two filters collected upstream of the CAOs showed that the presence of mineral dusts accounted for 30 % or more of the aerosol surface area. Considering the meteorology during the campaign and activity of the samples, it is likely that the majority of the INPs did not come from local sources but instead from long-range transport mechanisms more commonly associated with the Arctic haze phenomenon. As such, we suggest that the high INP concentrations measured during the ACAO campaign were due to the presence of biogenically enhanced dust, which dominated the ice nucleation processes, rather than sea-spray aerosol, in spite of the remoteness of the region.

These new measurements of INP concentrations measured in the ACAO campaign raise important questions for our understanding of Arctic mixed-phase clouds. The strength of the cloud-phase feedback depends on the balance between ice and liquid in clouds in the present day, with clouds with more ice having a greater negative feedback on climate in a warming world (Murray et al., 2021). Our results suggest that the magnitude of the cloud-phase feedback may be larger in Northern Hemisphere CAOs compared to their Southern Ocean equivalents, where it has been shown that the INP populations are many orders of magnitude smaller than in ACAO. Clearly, further work is needed to establish how representative ACAO was for CAO conditions in the Arctic. Nevertheless, future modelling work should investigate the response of Arctic mixed-phase clouds with these INP concentrations

to warming to reduce the uncertainty in this feedback.

The high INP concentrations measured in ACAO may also impact the transition between stratocumulus and cumulus clouds in CAOs, which is partially controlled by precipitation (Abel et al., 2017). Since enhanced INP concentration can enhance ice growth and thus precipitation, these high INP concentrations may act as a control on CAO development. Modelling that investigates the strength of the control that INP concentration exhibits on CAO development will be useful for understanding the impact of INPs on CAO cloud cover and thus radiative forcing. Finally, airborne measurements above and below the cloud deck have helped to establish the dominant INPs in these case studies. We recommend that more aircraft measurements be made in air that is of direct relevance to clouds in order to better understand the relationship between CAO cloud properties and INPs.

Chapter 3

Simulating clouds using the Unified Model and CASIM microphysics

Summary

CASIM is the double-moment microphysics module with interactive aerosol implemented by Field et al. (2023). It is used in regional configurations of the Met Office Unified Model (UM) and in large-eddy simulations using the Met Office-NERC Cloud model (MONC) (Brown et al., 2020; Field et al., 2023). The regional implementations include UKV, the Met Office's operational forecasting product for the United Kingdom (Bush et al., 2025). In this work, CASIM is used to simulate cold-air outbreaks observed during the Arctic Cold Air Outbreak campaign. In this chapter, I describe how CASIM operates and explain why it is an appropriate tool to model cold-air outbreaks.

3.1 Representation of hydrometeors

CASIM partitions cloud hydrometeors into two liquid species and three ice species. These species are liquid water (w), rain (r), ice crystals (i), snow (s) and graupel (g). In mathematical subscripts to follow, these species are referred to by their abbreviations. Additionally, the mass mixing ratio of water vapour, q_v is tracked. Liquid species are partitioned based on their size. Autoconversion (described later in Table 3.1) between liquid and rain is based on the mass-transfer autoconversion scheme of Khairoutdinov and Kogan (2000). The number transfer is calculated assuming a threshold diameter between the two species of 50 μm . Ice particles are partitioned based on their size and density. Snow particles are low density particles which grow through aggregation, while graupel particles are high-density particles which grow through riming (Field et al., 2023). The threshold diameter for autoconversion between ice and snow is 50 μm . This means that ice in CASIM is typically converted to snow shortly after nucleation.

CASIM is a **double-moment** microphysics scheme. Double-moment schemes prognose both the number concentration and mass mixing ratio of each species of hydrometeor (Morrison and Gettelman, 2008; Gettelman et al., 2010), whereas **single-moment** schemes prognose hydrometeor mass mixing ratio and use this to calculate number concentration based on assumptions about the particle size distribution. Number concentrations calculated this way are said to be **diagnosed** (Wilson and Ballard, 1999). The additional degrees of freedom associated with double-moment microphysics schemes means they are potentially able to produce outputs closer to observations than their single-moment counterparts (Field et al., 2023). However, double-moment schemes are more computationally expensive than single-moment schemes, and van Weverberg et al. (2014) notes that for the use of a second moment to provide significant improvements in model performance, other aspects of the model physics must perform well. To prognose number concentration and mass mixing ratio, CASIM represents the particle size distribution of each hydrometeor species ($\mathcal{N}_x$) using a generalised gamma distribution,

$$\mathcal{N}_x(D) = N_x \frac{\lambda_x^{1+\mu_x}}{\Gamma(1+\mu_x)} D^{\mu_x} \exp(-\lambda_x D), \quad (3.1)$$

where D is the particle diameter, μ_x is a slope parameter (2.5 for all species except snow, where $\mu_s = 2$) and λ_x is a shape parameter that is updated when the number and/or mass changes (Field et al., 2023). For each species, the number concentration N_x is calculated as the zeroth moment of the distribution (Whitby and McMurry, 1997; Khain and Pinsky, 2018). This integral is

$$N_x = \int_0^\infty \mathcal{N}_x(D) dD. \quad (3.2)$$

For all hydrometeors except snow, the mass mixing ratio, q_x is calculated as the third moment of the distribution,

$$q_x = \frac{\pi \rho_x}{6 \rho} \int_0^\infty \mathcal{N}_x(D) D^3 dD, \quad (3.3)$$

where ρ is the air density. For liquid and rain, ρ_x is the density of liquid water, but in the case of ice and graupel, ρ_x is the effective density, an empirically derived quantity that means ice can be approximated as spherical (Cotton et al., 2013). However, the effective density of snow aggregates is proportional to D^{-2} (Westbrook et al., 2004; Cotton et al., 2013), so the second moment of the distribution is used to calculate the mass mixing ratio of snow, such that

$$q_s = \frac{\rho_s}{\rho} \int_0^\infty \mathcal{N}_x(D) D^2 dD, \quad (3.4)$$

where $\rho_s = 0.026 \text{ kg m}^{-2}$ (Cotton et al., 2013; Field et al., 2023). Recently, triple-moment microphysics schemes have been developed. These use a third moment of the distribution¹ in combination with the number concentration and mass mixing ratio to calculate μ_x . Triple-moment schemes, though rarely deployed, are beginning to show improved performance

¹ not to be confused with the third moment of the distribution, used in Eq. 3.3 to calculate mass mixing ratio

on double-moment schemes (Milbrandt et al., 2021). However, the use of a third moment adds significant computational overheads.

When the CASIM module is called, the concentrations and mass mixing ratio of each of the five hydrometeors is adjusted according to the microphysical processes shown in Table 3.1. Full details of these processes can be found in Field et al. (2023).

Table 3.1: A summary of the microphysical processes included in CASIM (Field et al., 2023). Condensation and evaporation of cloud water is controlled outside CASIM by the bi-modal cloud fraction scheme of van Weverberg et al. (2021).

Process	Description	Hydrometeor
Accretion	Collection of a (usually) smaller hydrometeor by a (usually) larger hydrometeor.	cloud water by all others; ice by all but cloud water; rain by snow and graupel; snow by graupel
Activation	Activation of a cloud condensation nucleus into a droplet. By default, this is performed according to the Abdul-Razzak and Ghan (2000) scheme.	cloud water only
Aggregation	Self-collection of precipitation-size hydrometeors of a single species.	rain and snow
Autoconversion	Self-collection of a smaller species into the larger species of the same phase.	cloud water to rain; ice to snow
Deposition	Growth of ice particles by vapour deposition when air is supersaturated with respect to ice. Deposition ice nucleation is not included in this.	all frozen species
Evaporation	Evaporation rate of raindrops when entering air subsaturated with respect to water.	rain only
Ice nucleation (homogeneous)	Freezing of liquid droplets at temperatures below -38°C .	cloud water to ice; rain to graupel
Ice nucleation (heterogeneous)	Fully described in Section 3.2.1. By default, DeMott et al. (2010) is used when aerosol is processed and Cooper (1986) if not. Additionally, Bigg (1953) is used for the heterogenous freezing of raindrops, though this is contained within the homogeneous nucleation subroutine.	cloud water to ice

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Table 3.1 – continued from previous page

Process	Description	Hydrometeor
Melting	Melting of frozen species to become rain. Instantaneous for ice, but not for snow and graupel, which adhere to a parameterised melting rate.	ice to rain; snow to rain; graupel to rain
Secondary ice production	Increase in ice crystal concentration through the Hallett-Mossop process between -2.5°C and -7.5°C . The production rate is controlled by snow and graupel production rates.	all frozen species
Sedimentation	Species falling into lower model levels under gravity.	all species
Sublimation	Sublimation of frozen hydrometeors when entering air subsaturated with respect to ice.	all frozen species

While CASIM controls the microphysical processes within the cloud, the total water content and cloud fraction is controlled by the bimodal cloud scheme of van Weverberg et al. (2021), which considers the mixing of dry and moist air in entrainment zones at the boundaries of clouds.

3.2 Aerosol and its interactions with hydrometeors

CASIM can be coupled to a prognostic modal aerosol scheme such as the United Kingdom Chemistry and Aerosols model (UKCA) (Morgenstern et al., 2009; Mann et al., 2010; O'Connor et al., 2014) to specify the initial number concentration and mass mixing ratio of aerosols (Gordon et al., 2020; Field et al., 2023; Gordon et al., 2023). In this work, we use a new **two-way coupling** between UKCA and CASIM. Two-way coupling means that aerosol fields are passed to CASIM, modified by the cloud processes simulated in CASIM and returned to UKCA and the UM for further processing. These modified aerosol fields are then used in the next model timestep. In contrast, most previous work investigating aerosol-cloud interactions using CASIM has used **one-way coupling**, where aerosol fields from UKCA are passed to CASIM and inform the cloud microphysics, but no modifications are made to the aerosol fields by CASIM (Gordon et al., 2023).²

To couple UKCA to CASIM, aerosol is simplified to two types – soluble and insoluble – which are specified based on reanalysis data from the Copernicus Atmospheric Monitoring Service (CAMS). These are specified at the initiation of the run and at the boundaries of the domain,

²A more detailed discussion of previous work with CASIM is in Section 3.3.

where they may be advected in or out of the domain depending on the wind direction, and can be overwritten to represent measurements. When not contained in a hydrometeor, the mass mixing ratio and number concentration of soluble aerosol is traced across three size modes: Aitken, accumulation and coarse; while the same properties of insoluble aerosol are traced across the accumulation and coarse modes. These aerosols can activate hydrometeors, and by default this is performed according to Abdul-Razzak and Ghan (2000).

Within the CASIM module, 20 tracers (numbers which store information through different model processes) are used to track the total mass and number of both soluble and insoluble aerosol contained in each of the five species of hydrometeor. When hydrometeors undergo some of the microphysical processes shown in Table 3.1, the aerosol populations associated with these hydrometeors are adjusted accordingly. These adjustments are described in detail in Table 3.2.

For insoluble aerosol, both mass and number are conserved during the CASIM timestep unless they are sedimented out of the domain. For instance, if ice crystals containing insoluble aerosol accrete liquid droplets containing insoluble aerosol producing snow, the number of insoluble aerosol in the newly formed snow will be the sum of the insoluble aerosol numbers in the ice and the droplets. In contrast, for soluble aerosol, each hydrometeor has one single aerosol, meaning that aerosol number is not conserved, while aerosol mass is conserved. This means that evaporation of large hydrometeors that have grown through accretion or aggregation releases fewer, larger aerosol than entered the smaller hydrometeors that they grew from (Miltenberger et al., 2018). Dry deposition and scavenging of aerosol occurs outside the CASIM module and is handled by UKCA (Mann et al., 2010; Jones et al., 2022).

Table 3.2: CASIM microphysical processes that affect aerosol tracer values. Aggregation, autoconversion and SIP do not affect the number or mass of aerosol in a given tracer — for autoconversion, the quantity of aerosol in each hydrometeor species is diagnosed at each timestep based on mass partitioning. Additionally, deposition does not result in changes to aerosol since the deposition process does not include nucleation of new ice crystals by deposition.

Process	Effect on aerosol tracers
Accretion	Aerosol mass is transferred from the collected species to the collector.
Activation	Aerosol mass and number are transferred from the soluble accumulation mode tracer and the insoluble coarse mode tracer to the soluble/insoluble aerosol in liquid tracers.

Continued on next page...

Table 3.2 – continued from previous page

Process	Effect on aerosol tracers
Evaporation	For soluble aerosol, aerosol mass is transferred to the accumulation or coarse mode tracer, depending on the aerosol average size that is diagnosed. For insoluble aerosol, aerosol mass in liquid is transferred to the coarse mode tracer. For the insoluble aerosol only, the number of activated aerosol is transferred to the coarse mode tracer when ice nucleation is based on aerosol.
Ice nucleation	Depending on the parameterisation, aerosol mass and number is transferred from at least one of the the insoluble coarse mode, soluble accumulation mode or aerosol in liquid tracers to the appropriate tracers for aerosol activated in ice. In the default aerosol-aware configuration, soluble aerosol does not nucleate ice.
Melting	For soluble and insoluble aerosols, aerosol mass in ice is transferred to the soluble/insoluble aerosol mass in liquid tracers.
Sublimation	For soluble aerosol in ice, aerosol mass and number is transferred to the soluble accumulation mode tracers. For insoluble aerosol in ice, aerosol mass and number is transferred to the insoluble coarse mode tracers.
Sedimentation	Aerosol mass and number contained in hydrometeors that have reached the surface are removed.

Though the number and mass of each aerosol type in each hydrometeor species is traced within CASIM, the parent model (the UM) does not have the capacity to trace these quantities owing to the large computational overhead associated with this and the requirement to be compatible with UKCA and other UM code. Accordingly, when the CASIM module returns data to the UM for the rest of the model timestep, the twenty tracers of aerosol in hydrometeors are reduced to seven or eight depending on the model configuration. These are shown in Table 3.3, alongside the tracers for aerosol outside hydrometeors.

Table 3.3: Aerosol tracers in the UM which are coupled to CASIM. Here, “liquid” and “ice” refer to all liquid and frozen species unless using the asterisked optional case (not used in this work) where aerosol in cloud water and rain are treated separately.

	Tracers for aerosol outside hydrometeors		Tracers for aerosol activated in hydrometeors	
	Mass	Number	Mass	Number
Soluble	Aitken	Aitken	in all liquid/in cloud water*	total
	accumulation	accumulation	in rain*	
	coarse	coarse	in all ice	
Insoluble	accumulation	accumulation	in all liquid	in all liquid
	coarse	coarse	in all ice	in all ice

3.2.1 Aerosol-aware ice nucleation

In CASIM, ice nucleation can be **passive**, where the INP concentration (and thus the freezing of droplets) is diagnosed based on temperature and meteor concentrations, or **interactive**, where the INP concentration is prognosed from aerosol, and aerosol is moved from the in-droplet tracer to the in-ice tracer for further processing.

When ice nucleation is passive, the default ice nucleation scheme is the Cooper (1986) parameterisation for primary ice crystal concentrations. This is **aerosol-unaware** and depends only on temperature. The parameterisation is

$$N_{\text{INP}} = F_{\text{Cooper}} \exp[-a(T_0 - T)], \quad (3.5)$$

where $F_{\text{Cooper}} = 5 \text{ m}^{-3}$, $a = 0.304 \text{ K}^{-1}$ and T_0 is the triple-point of water, 273.15 K (Cooper, 1986; Huang et al., 2025). Using this scheme, the expected number of primary ice crystals for a grid box of a given temperature is calculated. If $N_i < N_{\text{INP}}$, droplets are frozen to reach N_{INP} , depleting N_w and q_w , such that

$$\frac{dN_i}{dt} = \frac{\min(N_w, N_{\text{INP}} - N_i)}{\Delta t}, \quad (3.6)$$

where Δt is the model timestep and the minimum function prevents more droplets being depleted than available in the grid box. If CASIM is run using a fixed droplet number concentration, the value of N_{INP} according to the Cooper parameterisation is instead used to nudge the ice crystal concentration back towards climatological values,

$$\frac{dN_i}{dt} = \frac{\tau(N_{\text{INP}} - N_i)}{\Delta t}, \quad (3.7)$$

where $\tau = 0.8$ is a relaxation constant that moderates the strength of the nudging. This can remove ice crystals above the number that would be expected, preventing a potential infinite source of ice crystals.

Rather than depending solely on environmental variables such as temperature, passive ice nucleation can be **aerosol-aware**, where either the soluble or insoluble aerosol tracers (or both) provide a limit on the ice nucleation rate. By necessity, aerosol-aware schemes are used when ice nucleation is interactive. The default aerosol-aware ice nucleation scheme in CASIM is an adapted version of the DeMott et al. (2010) parameterisation for INP concentration and makes the assumption that the coarse-mode insoluble aerosol represents dust. To do this, the number of contact-INPs, $N_{\text{INP, con.}}$, is calculated from the number of coarse-mode insoluble aerosol in air ($N_{\text{insol, a}}$),³ while the number of immersion-INPs, $N_{\text{INP, imm.}}$, is calculated from the number of insoluble aerosols contained in liquid droplets, $N_{\text{insol, w}}$. For a grid-box of temperature T , these are

$$\begin{aligned} N_{\text{INP, con.}} &= a (T_0 - T)^b \eta_c \cdot (N_{\text{insol, a}})^{[c(T_0 - T) + d]} \\ N_{\text{INP, imm.}} &= a (T_0 - T)^b \eta_i \cdot (N_{\text{insol, w}})^{[c(T_0 - T) + d]}, \end{aligned} \quad (3.8)$$

Here, $\eta_c = 1 \times 10^{-4}$ and $\eta_i = 1$ are scale parameters for the efficiency of contact and immersion nucleation, respectively, and the insoluble aerosol concentrations are measured in m^{-3} . The fit parameters are $a = 5.94 \times 10^4$, $b = 3.33$, $c = 2.64 \times 10^{-2} \text{ K}$ and $d = 3.33 \times 10^{-2}$. As in the case of the passive Cooper parameterisation, physical limitations are applied to stop unrealistic ice concentrations. The number of potential ice nuclei is limited by the number of droplets available to freeze, regardless of whether nucleation occurs via the immersion or contact modes. Additionally, no more than $\chi = 90 \%$ of the coarse mode insoluble aerosol in air may nucleate ice in a single timestep, such that

$$\frac{dN_i}{dt} = \min [N_{\text{INP, imm.}} + \max (N_{\text{INP, con.}}, \chi N_{\text{insol, a}}), N_w] (\Delta t)^{-1} \quad (3.9)$$

After nucleation has occurred, the number and mass of insoluble aerosol causing is transferred from the coarse mode tracer to the in-ice tracer.

The ice nucleation scheme can be adjusted to reflect other DeMott-like parameterisations which relate aerosol number concentration to INP concentration. Alternatively, schemes which relate aerosol surface area to INP concentration, such as the Niemand et al. (2012) can be chosen, where

$$N_{\text{INP, imm.}} = (N_{\text{insol, w}}) \exp [-a (T_0 - T)] \cdot 4\pi (\bar{r}_{\text{insol}})^2 \cdot \exp \left[2 (\ln \sigma_{\text{insol}})^2 \right]. \quad (3.10)$$

Here, the total surface area of the insoluble aerosol is calculated using the mean radius of the coarse insoluble aerosol mode, $\bar{r}_{\text{insol}}$ and its modal width σ_{insol}^2 . The fitting parameter $a = 0.517$. Contact nucleation is not considered when using the Niemand scheme, while the physical limitations in Eq. 3.9 apply. As in the DeMott scheme, insoluble aerosol is trans-

³To avoid verbosity, the subscript *insol* will, unless otherwise specified, refer only to insoluble aerosol in the coarse mode. This is because insoluble aerosol in the accumulation mode does not interact with any of the CASIM processes and concentrations of accumulation-mode aerosol are orders of magnitude lower than concentrations of coarse-mode aerosol in the simulations described in Chapter 4 and Chapter 5.

ferred from the coarse mode tracer to the in-liquid tracer once nucleation has occurred.

The terms aerosol-aware, aerosol-unaware, interactive and passive will be used throughout this work. To aid the reader, their definitions are compiled in Table B.1.

3.3 Using CASIM to study aerosol-cloud interactions

CASIM is a double-moment microphysics scheme that has been used to study cloud microphysics and aerosol-cloud interactions in a wide range of meteorological conditions and strongly contrasting regions on Earth. These include Sahelian mesoscale convective systems (Maybee et al., 2025), natural and contrail cirrus (Baran et al., 2025; Zhang et al., 2025), marine stratocumulus (Sansom et al., 2024) and cumulus (Spill et al., 2019) and deep convection (Hawker et al., 2021; Finney et al., 2025).

Previous research has shown that CASIM is able to represent mixed-phase clouds similar to the CAO clouds studied in this work reasonably well, though these studies have only used passive ice nucleation and most have used aerosol-unaware ice nucleation schemes. For instance, Huang et al. (2025) studied two CAOs at different temperatures in the Labrador Sea. When comparing a CAO with similar cloud temperatures to those in the ACAO campaign (Section 2.2.1) to satellite observations, they found a slight overestimate of ice water content (IWC) and an aligned slight cold bias in cloud-top temperature. Similarly, a study of mixed-phase frontal cloud off the Antarctic coast by Smith et al. (2025) showed that CASIM captured liquid water content (LWC) well (if slightly underestimating it) but produced too much snow relative to observations. However, changing the INP parameterisation to one more representative of the region reduced the high bias in snow concentration. Neither of these studies used aerosol-aware ice nucleation, and both specified droplet number rather than using an activation scheme such as Abdul-Razzak and Ghan (2000). In contrast, Price et al. (2025) simulated summertime mixed-phase Antarctic clouds over the Southern Ocean using CASIM coupled to UKCA, allowing aerosols to activate into droplets (though they used the aerosol-unaware Cooper ice nucleation scheme). They found that the default UKCA aerosol concentration was too low, resulting in reduced LWC relative to observations, but when fixing the cloud droplet number to a value consistent with observations, this was significantly improved. This suggests that aerosol activation in CASIM behaves as expected.

There have been fewer studies utilising CASIM's ability to trace aerosol through cloud processes. Considering a case study of convective clouds over South-West England, Miltenberger et al. (2018) compared CASIM with interactive aerosol processing to an implementation of CASIM with passive aerosol-aware processing, where droplet activation and ice nucleation were diagnosed from aerosol but none of the aerosol processes in Table 3.2 were performed. In the interactive case, cloud-base aerosol was depleted relative to the passive case through scavenging, representing a more realistic simulation of the cloud system. This enabled the authors to investigate the sensitivity of convective cell size to aerosol. Ad-

ditionally, (Field et al., 2023) used CASIM coupled with two different single-moment aerosol schemes: MURK, a submodule of the UKV model (Clark et al., 2008) in a mid-latitude case and ARCL, a global climatology (Bellouin et al., 2013) in a tropical case. They found evidence that one-way coupling of aerosol to CASIM improved cloud albedo relative to CASIM uncoupled to an aerosol model by producing more realistic droplet numbers. However, they noted that this did not hold for cirrus clouds.

In this work, we will use CASIM's full suite of aerosol microphysics to investigate the role of INPs cold-air outbreak clouds. Using an aerosol-aware, interactive ice nucleation scheme with CASIM coupled two-way to UKCA allows us to investigate the effect of INP as a limiting factor on ice nucleation, as aerosol which has nucleated ice is unable to further nucleate, and may even be removed from the system.

Chapter 4

Evolution of ice-nucleating particle concentration in an Arctic cold-air outbreak

Abstract

INPs strongly affect the properties of mixed-phase cloud systems such as CAOs, but measurements of INP concentration in CAOs made in Chapter 2 suggested that INPs may be efficiently removed during CAOs. However, calculations of cloud-phase feedback use cloud models with a fixed or aerosol-unaware INP concentration. If the removal of INPs is strong, these modelling choices may be inappropriate since they will not represent the change in INP concentration across the domain. To assess the impact of aerosol removal on INP concentrations in CAOs, we used a new set-up of the Met Office Unified Model in its regional configuration that couples two-moment microphysics to interactive aerosol processing to simulate a case study where INP removal was observed. We found that INPs are processed and removed during CAOs, though the rate of INP removal was smaller than suggested by measurements. However, INP removal had a negligible impact on INP concentrations in the cloud, which were predominantly controlled by cloud temperature.

4.1 Introduction

In Chapter 2, we presented several measurements of INPs made in the vicinity of CAO clouds over the Norwegian and Barents seas. These INP concentrations were among the highest previously measured in the Arctic, with high concentrations of INPs most prominent in CAOs with predominantly northerly flows. The INPs measured had active site densities similar to previously reported measurements of mineral dusts with ice-active biogenic components. Additionally, the highest active site densities were found above the cloud, suggesting that

the mineral dust, rather than sea spray aerosol, dominated the active INPs, while scanning electron microscopy also revealed that mineral dusts contributed approximately 30% of the total aerosol surface area.

Due to an apparent lack of local sources of this mineral dust, it was hypothesised that the main source of INPs measured in CAOs during the ACAA campaign was long-lived fertile dust aerosols that had undergone long-range transport to the high Arctic through mechanisms associated with Arctic haze (Section 2.4). As a CAO airmass is ejected from the Arctic, these aerosols were thought to be ejected also, and responsible for primary ice production in the CAO clouds. Once in the cloud system, these long-lived INPs ejected from the Arctic may then control the ice processes in the cloud. However, this is a two-way interaction. If an ice particle nucleated by an INP grows to a size which is large enough to precipitate, it will be removed, reducing the number concentration of INPs. Additionally, immersion-mode INPs such as dust act as CCN prior to nucleating ice (Rosenfeld et al., 2001; Kumar et al., 2009; Karydis et al., 2017). These INPs may be removed from the cloud system through other mixed-phase microphysics, such as being incorporated into a droplet which is later incorporated into an ice particle through a collision, causing the resultant hydrometeor to reach a size where precipitation is possible.

On multiple occasions during the ACAA campaign, INP concentrations were observed to reduce downstream in CAOs by between 30 % and 70 % (as described in Section 2.3.5), suggesting that INPs may be removed by the cloud system as it develops. However, this contradicted findings from remote sensing experiments which hypothesised that INP concentrations in CAOs increased downstream due to the addition of sea spray aerosol (Section 2.4). As discussed in Section 1.4, a decline in INP concentration during the transit of air through a CAO due to the removal of aerosol could result in differences in the downstream cloud albedo relative to a CAO with a constant INP concentration. This could then result in modifications to the strength of the cloud-phase feedback in the clouds downstream. Accordingly, understanding the changes in INP concentration through a CAO is important, since cloud modelling typically uses simplified aerosol-unaware representations of INPs. Even when ice nucleation is aerosol-aware, it is rarely interactive, meaning that the freezing initiated by INPs cannot result in their removal (Section 1.2.5 and Section 3.3). As such, if the removal of INPs is strong, evaluations of the cloud-phase feedback using these methods could prove to be inaccurate.

In this chapter, we present modelling of a case study from the ACAA campaign where the INP concentration was observed to reduce downstream through the outbreak. To do this, we use CASIM, a two-moment microphysics module with interactive aerosol which has a new two-way coupling to UKCA. As described in Chapter 3, this allows us to trace the concentration of INPs through the CAO, while allowing INPs to be removed by aerosol processing within clouds. We first evaluate the ability of CASIM to simulate CAOs using interactive droplet activation, ice nucleation, in-cloud processing of aerosol and a two-way coupling to

UKCA. We then use these results to ask:

- to what extent are INPs removed during a CAO which evolves over the period of about two days?
- how does the cloud-relevant INP concentration vary through a CAO?

4.2 The case study: flight c280

To investigate the removal of INPs in CAOs, we have chosen a CAO on 29 March 2022 which was measured by flight c280 of the ACAO campaign. Figure 4.1a and Figure 4.1b show the structure of the flight. After take-off from Kiruna, Sweden, at 07:58 UTC, the aircraft transitioned to approximately 74.5°N 20.5°E at 09:29 UTC. After this, the aircraft began a 20 min “straight and level” aerosol sampling run (c280r1) in the opposite direction to the prevailing wind at altitude 1570 m, above the cloud top and over the marginal sea ice. The aircraft then turned 180°, descended to sea level and began a second “straight and level” with the prevailing wind at 327 m above the marginal ice zone for 15 min. Following this sampling run, the aircraft continued to travel with the prevailing wind for approximately 15 min, while performing a vertical profile of the cloud by ascending to cloud-top, descending to cloud-bottom, and ascending to cloud-top again. At cloud top, the aircraft turned and performed a third, 23 min “straight and level” approximately perpendicular to the prevailing wind (c280r3) at altitude 2010 m, turning halfway through in order to return to the original path. After a further descent in the direction of the prevailing wind to profile the cloud further, a final 20 min “straight and level” (c280r4) was performed approximately perpendicular to the prevailing wind. During this run, some deviations were made to the straight path to avoid pockets of precipitation that may have led to destruction of the aerosol filter. Finally, at 11:25 UTC, the aircraft made a final ascending profile through the cloud before transiting back to Kiruna.

In addition to the four measurements of INPs during the flight, measurements of INP concentration were made using further downstream at a hilltop measurement station at Andenes (location shown on Figure 4.1a). These measurements were made using a different technique, where samples of aerosol were collected by a liquid impinger and INP concentrations were measured using a droplet freezing assay (David et al., 2019; Gjelsvik et al., 2025). Using the HYSPLIT trajectory model, these INP measurements were timed to measure the same air mass that had been measured upstream (David, et. al, 2022–2025; personal communication).

Figure 4.1c shows that cloud measurements were made at temperatures ranging from 255 K to 268 K, and were rarely in the temperature range associated with the Hallett-Mossop SIP process, the only SIP process included in CASIM. However, the HM process may have been influential at the southern extent of the clouds, while other SIP processes which are not represented in CASIM may have been present in reality. Where measurements were made in-

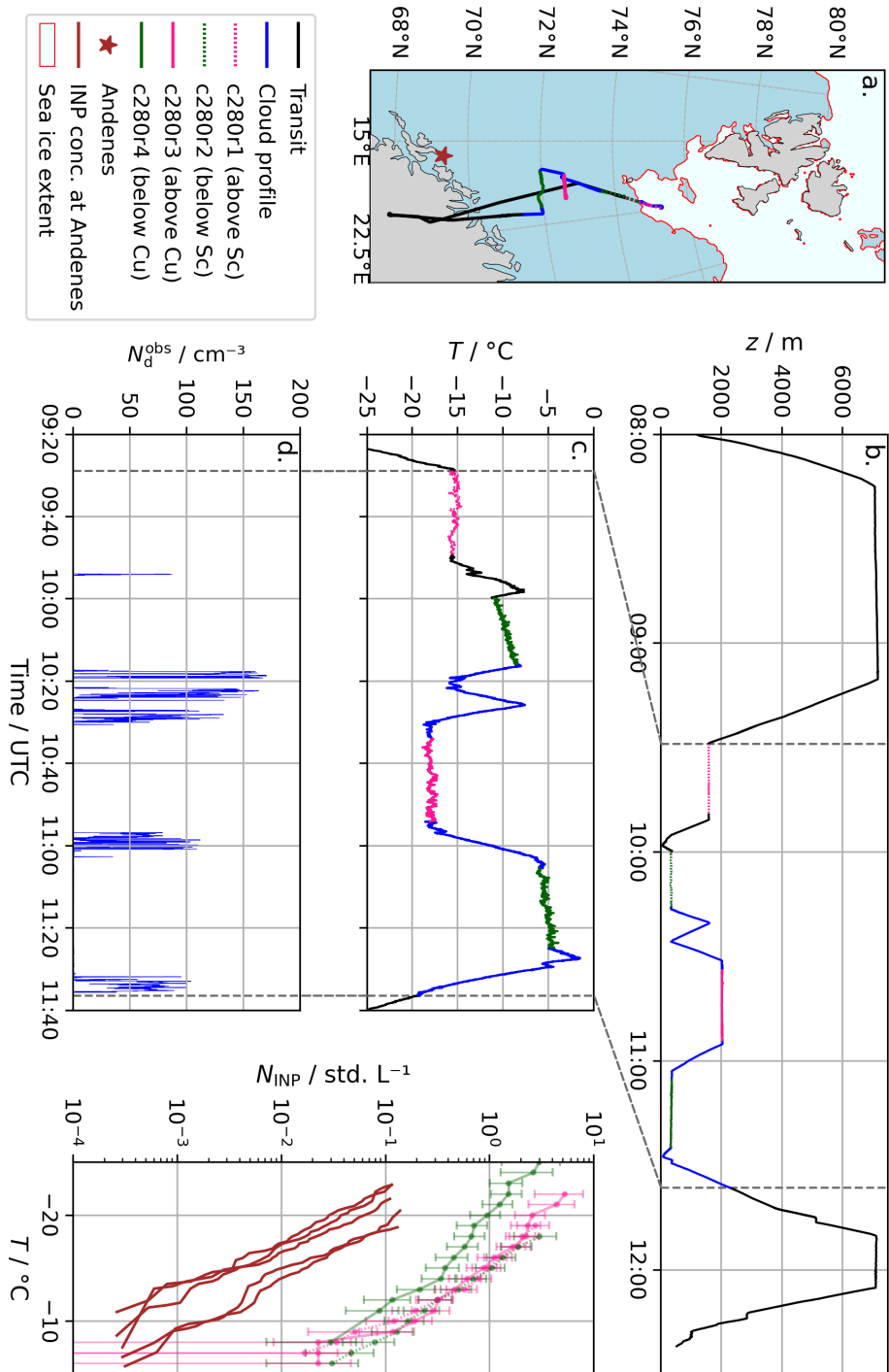


Figure 4.1: Aircraft instrument data from flight c280. Panels (a) and (b) show the aircraft flight path and altitude during the transit, cloud profile and aerosol filter sampling legs. Panel (c) shows the temperature measured by the non-deiced Rosemont probe, while (d) shows the cloud droplet number concentration measured by the CDP during cloud profiles. Panel (e) shows INP concentrations measured during each leg, as previously described in Chapter 2, compared to measurements of the same air masses at Andenes (David, et. al, 2022; personal communication).

cloud, peak liquid droplet number concentrations, N_d^{obs} , ranged from 100 cm^{-3} to 170 cm^{-3} , as shown in Figure 4.1d.

Figure 4.1e shows the INP concentrations measured during this flight and the INP concentrations measured at Andenes. There is little difference in INP concentration between the above-cloud upstream measurement, the downstream above-cloud measurement and the upstream below-cloud measurement. In contrast, there is nearly a five-fold reduction in INP concentrations in the downstream below-cloud measurement. There is a further 10 to 100-fold reduction in INP concentrations between the aircraft INP measurements and the ground-based INP measurements made at Andenes. This could suggest a strong removal of INPs, but as discussed in Section 2.2.2, there is considerable uncertainty when comparing the results of different INP measurement procedures that may account for a large proportion of this discrepancy.

4.3 Model setup

In this work, version 13.8 of the UM (Brown et al., 2012) was used with Regional Atmosphere and Land (RAL) configuration 3.3 (Bush et al., 2025) with a regional nesting suite to model the CAO measured by flight c280 of the ACAO campaign. The nested suite inherited boundary conditions every hour from archived Global UM analysis run with resolution N680 with Global Atmosphere 8.0 and Global Land 9.0 configurations (Willett et al., 2026). At N680 resolution, the global model uses grid cells of 0.26° longitude and 0.18° latitude, resulting in an approximately isotropic grid spacing of 19 km at 50° N (Walters et al., 2017). The nesting suite had 800 grid points separated by 0.018° (approximately 2 km) in both latitude and longitude, centred on $75^\circ \text{ N } 24^\circ \text{ E}$. To maintain an approximately isotropic grid, a “rotated pole” configuration was used, whereby the equator and prime meridian of the grid passes through the central point of the nest (Bush et al., 2025). The nesting suite had 70 vertical levels extending to 40 km above ground level, with a finest vertical resolution of 17 m at the surface and vertical resolutions of order 150 m between 1000 m and 2200 m. For this case study, the model was initialised at 1200 UTC on 28 Mar 2022, the day before flight c280 during ACAO. The model timestep was 60 seconds and the model was run for 24 hours, concluding at 1200 UTC on 29 Mar 2022, after measurements had ceased. Sea ice used in the model was taken from the daily Operational Sea Surface Temperature and Ice Analysis (OSTIA) product.

Interactive aerosol-cloud microphysics was performed by the CASIM module coupled to UKCA, described fully in Chapter 3. However, modifications to the aerosol fields and ice nucleation scheme were made to represent the c280 case study. The accumulation-mode soluble and coarse-mode insoluble aerosol fields were initialised according to profiles are shown in Figure 4.2a. To derive the profiles, a profile of total aerosol number concentration was calculated from the PCASP and CDP measurements (Section 2.2.4). Aerosol concentrations were averaged on each model level, which were then smoothed using a rolling aver-

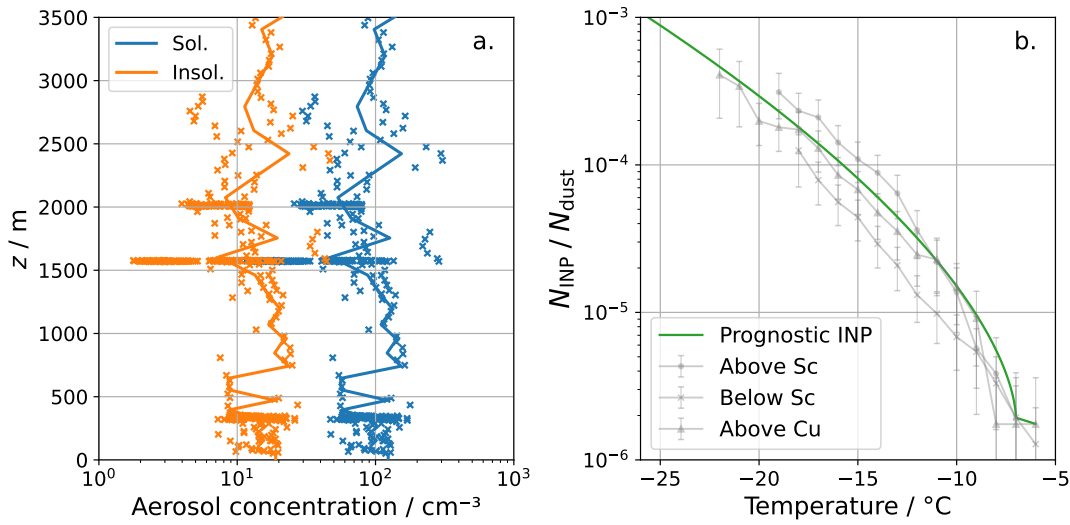


Figure 4.2: Panel (a): aerosol profiles used by CASIM. Concentrations of aerosol with diameters between $0.1\ \mu\text{m}$ and $10\ \mu\text{m}$ measured by the PCASP and CDP probes during flight c280, and the aerosol profile derived from them. Each cross depicts the average aerosol number measured over a 10-second period, excluding time over the Scandinavian peninsula. The ratio of soluble to insoluble aerosol was derived from scanning electron microscopy measurements. Panel (b): the INP parameterisation used by the model, Eq. 4.1.

age (with varying window size) to balance a faithful replication of the aerosol measurements and the reduction of discontinuities in the aerosol profile. There were no composition measurements available on the day of the flight. Given that the INP concentrations associated with CAOs with strong northerly components throughout the campaign varied by less than an order of magnitude and assuming that an upstream reservoir of long-lived INPs will be well-mixed, we have assumed that SEM analysis of filter samples made in similar conditions would approximately represent the relative composition of the aerosol. Accordingly, the proportion of upstream aerosol which SEM analysis identified as dust, $\alpha = 0.133$, was calculated. The total aerosol concentration was then scaled by α and $1 - \alpha$ to partition aerosol into soluble and insoluble types. The aerosol fields were initially horizontally homogeneous, and the profiles were used to resupply aerosol from the lateral boundaries every hour. Aitken- and coarse-mode soluble aerosol were taken from CAMS reanalysis data, as was accumulation-mode insoluble aerosol, though none of these aerosol modes interacted with CASIM.

The default INP parameterisation (Section 3.2.1) was also replaced with a parameterisation to represent the case study. This parameterisation is shown in Figure 4.2b. Since we concluded in Section 2.5 that fertile dusts were the dominant source of INPs during ACAO, this parameterisation acted on insoluble aerosol only. To calculate the INP parameterisation, the above-cloud $N_{\text{INP}}/N_{\text{aer}}$ measurements were scaled by α and a four-parameter function of the form used in Section A.3 was fitted to their average. The above-cloud measurements were selected to represent the cloud system prior to removal processes, since these were unlikely to have been affected by cloud processing. Ice nucleation was assumed to be

Table 4.1: Values of parameters for the INP parameterisation used in the model given by Eq. 4.1.

Parameter	a	b / K^{-1}	c / K	d / K^{-b}	$e / 10^{-7}$	$f / 10^{-8} \text{K}^{-1}$	g / K
Value	-15.171	1.050	266.15	0.610	2.33	2.4	267.15

immersion-mode, and only aerosol contained in water (with concentration $n_{\text{insol}, \text{w}}$) could freeze. Since the best-fitting curve for the average substantially underestimated ice nucleation between -6°C and -7°C , a taper was applied to the fit in this temperature range. No freezing was permitted at $T > -6^\circ\text{C}$, the highest temperature at which freezing was experimentally observed. As such, the parameterisation was

$$N_{\text{INP}} = \begin{cases} \alpha^{-1} \exp \left[a + b(c - T)^d \right] N_{\text{insol}, \text{w}}, & T < -7^\circ\text{C} \\ \alpha^{-1} (e + f(T - g)) N_{\text{insol}, \text{w}}, & -7^\circ\text{C} \leq T < -6^\circ\text{C} \\ 0, & T > -6^\circ\text{C}. \end{cases} \quad (4.1)$$

where $a, b \dots g$ are parameters of the fit given in Table 4.1.

Akin to Eq. 3.9, the number of ice nuclei was limited by the number of droplets available, such that

$$\frac{dN_i}{dt} = \frac{\min(N_{\text{INP}}, N_{\text{insol}, \text{w}})}{\Delta t}. \quad (4.2)$$

4.4 Results

4.4.1 Comparing modelled clouds to in-situ and satellite measurements

Figure 4.3a and Figure 4.3b show that the cloud structure in the model compares well with data from the MODIS instrument. The development of cloud occurs in the same places in the model and reality, with noticeable clear air off the west coast of Spitsbergen and reduced cloud cover immediately up and downwind of Bjørnøya¹. The cloud formation is pinned to the sea ice, for which modelled sea ice using OSTIA and observations made by MASIE also compare well. Downstream in the CAO, cumulus cells are of the same size in the model and satellite imagery, while upstream, cloud-street width is well-represented in the model when wider than the 2 km resolution of the model. However, immediately downstream of Bjørnøya (box 4), cloud streets visible in the satellite imagery are unresolved in the model — this may be due to the coarseness of the model resolution. Considering cloud albedo, Figure 4.3b (the model) and Figure 4.3d (radiometry data) show similar features, though the albedo is lower in the satellite data than the model at the southwest extent of the outbreak. This is likely to be due to the averaging of cloud-free and cloudy air in the

¹Bear Island

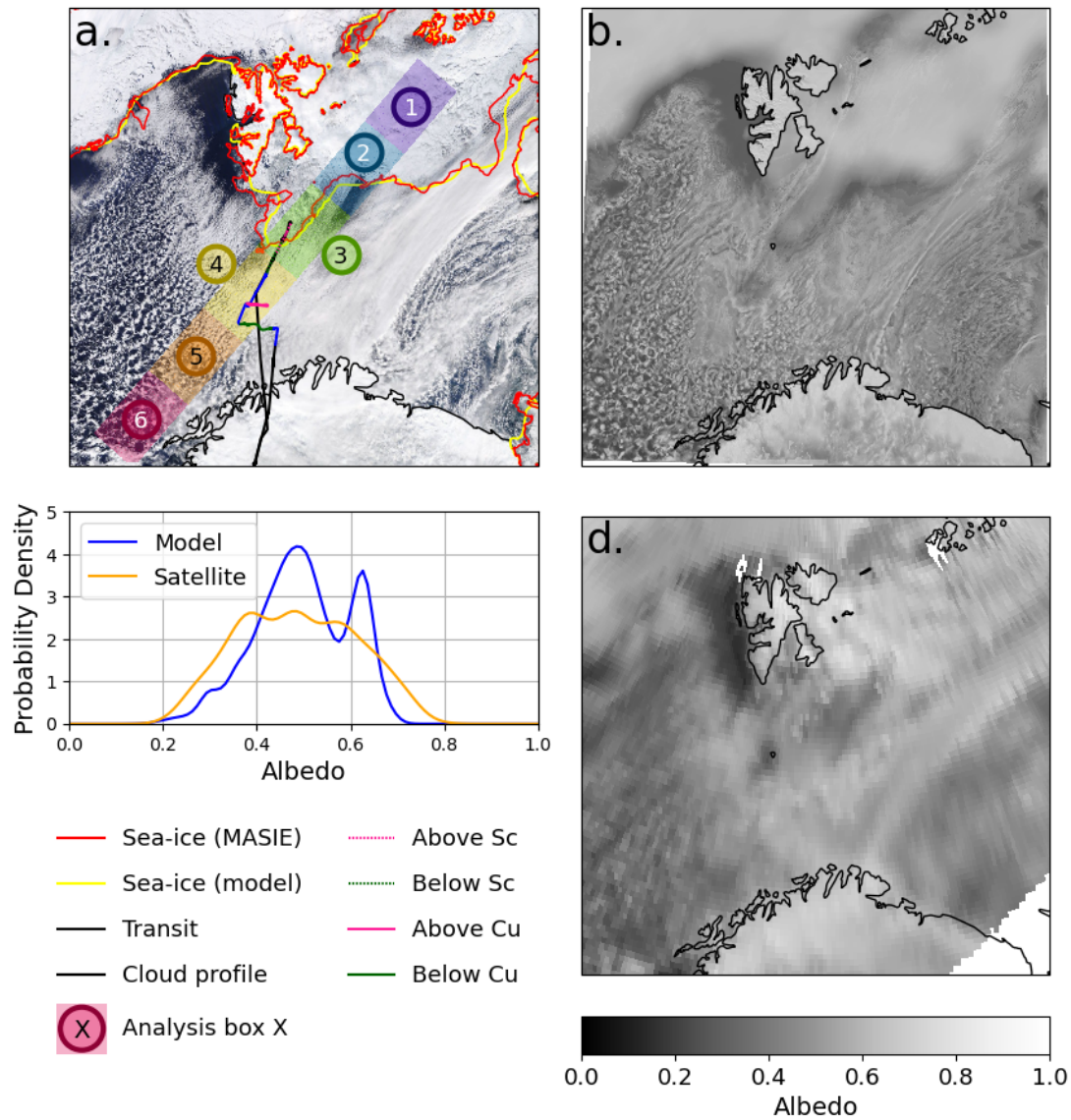


Figure 4.3: A comparison of cloud structure and albedo between the model and satellite observations at approximately 11:00 UTC on 29 March 2022. Panel (a) shows real-colour satellite imagery from Terra MODIS (MODIS Land Science Team, 2020). Superimposed is the track of flight c280, and the sea ice boundary from MASIE observational data, which is compared to the OSTIA ancillary sea ice used in the model. Atop the figure are six coloured boxes which correspond to regions used to take average profiles and column integrals in later analysis. These boxes are in the direction of the mean wind flow, which is from box 1 to box 6. Panel (b) shows the scene albedo according to the model, while panel (c) shows scene albedo calculated from top-of-atmosphere shortwave radiative fluxes measured by the Clouds and the Earth's Radiant Energy System (CERES) radiometer on the Terra satellite (Su et al., 2015). Panel (d) shows a probability density function of scene albedo for both the model and CERES compared over the area covered by all coloured boxes.

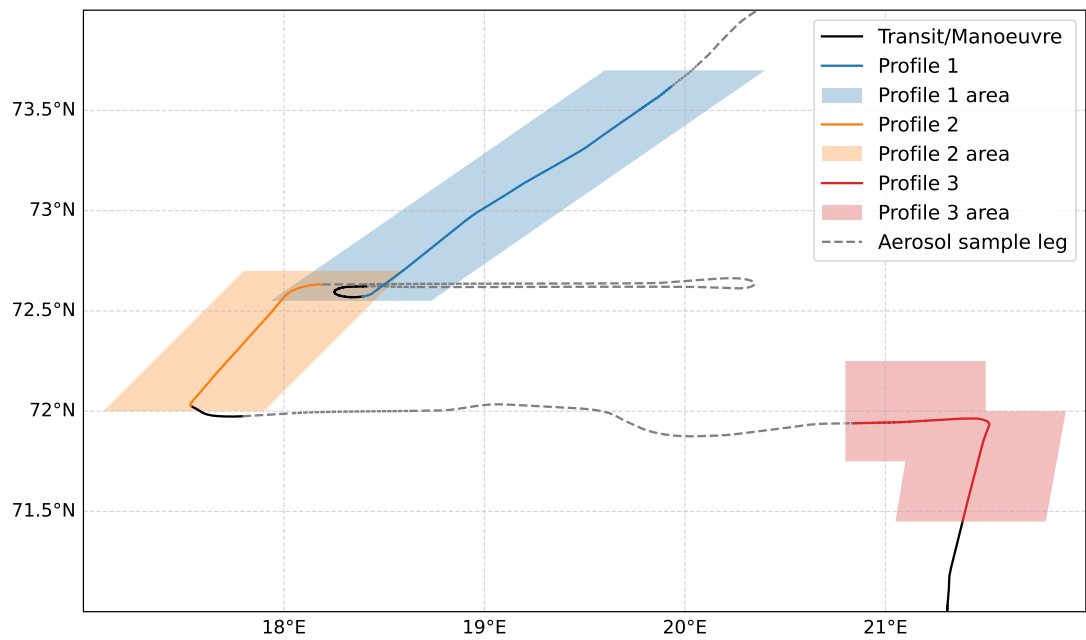


Figure 4.4: Locations of cloud profiles performed during flight c280. Shaded areas depict the regions over which model profiles were taken. Profiles are shown at a larger scale than Figure 4.1a, which shows the location of these relative to Svalbard and the Scandinavian peninsula, and Figure 4.3a, which shows their locations relative to the cloud system as shown in satellite imagery.

lower-resolution satellite data; this is reflected in the probability density functions (PDFs) of the region of interest shown in Figure 4.3c. Here, the PDF of modelled albedo has a two-peaked distribution from 0.4 to 0.7, while the PDF of measured albedo has a single, flat peak. The two peaks of the model albedo represent the lighter and darker regions of the cloud streets which are averaged by the satellite.

To assess the ability of the model to capture cloud microphysical properties, we can compare the modelled liquid and ice contents to three cloud profiles made by the aircraft. As shown in Figure 4.3a and Figure 4.3b, these profiles were taken in the region where the model did the worst job of capturing the macrophysical structure and should represent a lower bound of model fidelity. The nature of the open and closed cells in the CAO means that even small dynamical differences between the model and reality will result in modelled clouds being not exactly coincident with those observed. Accordingly, to make comparisons of microphysical quantities, comparing directly to the nearest model grid box is often uninformative. Consequently, we compare aircraft profiles to the average across an area spanning 0.4° approximately perpendicular to the direction of the aircraft (21 to 23 grid cells). These areas are shown in Figure 4.4, while comparisons of the model profiles to the aircraft profiles are shown in Figure 4.5. For reference, horizontal winds and relative humidity are shown in Figure C.1 while direct comparisons between the model and the nearest grid box are shown in Figure C.2.

The cloud droplet number was measured by the CDP (Rosenberg et al., 2012), while liquid

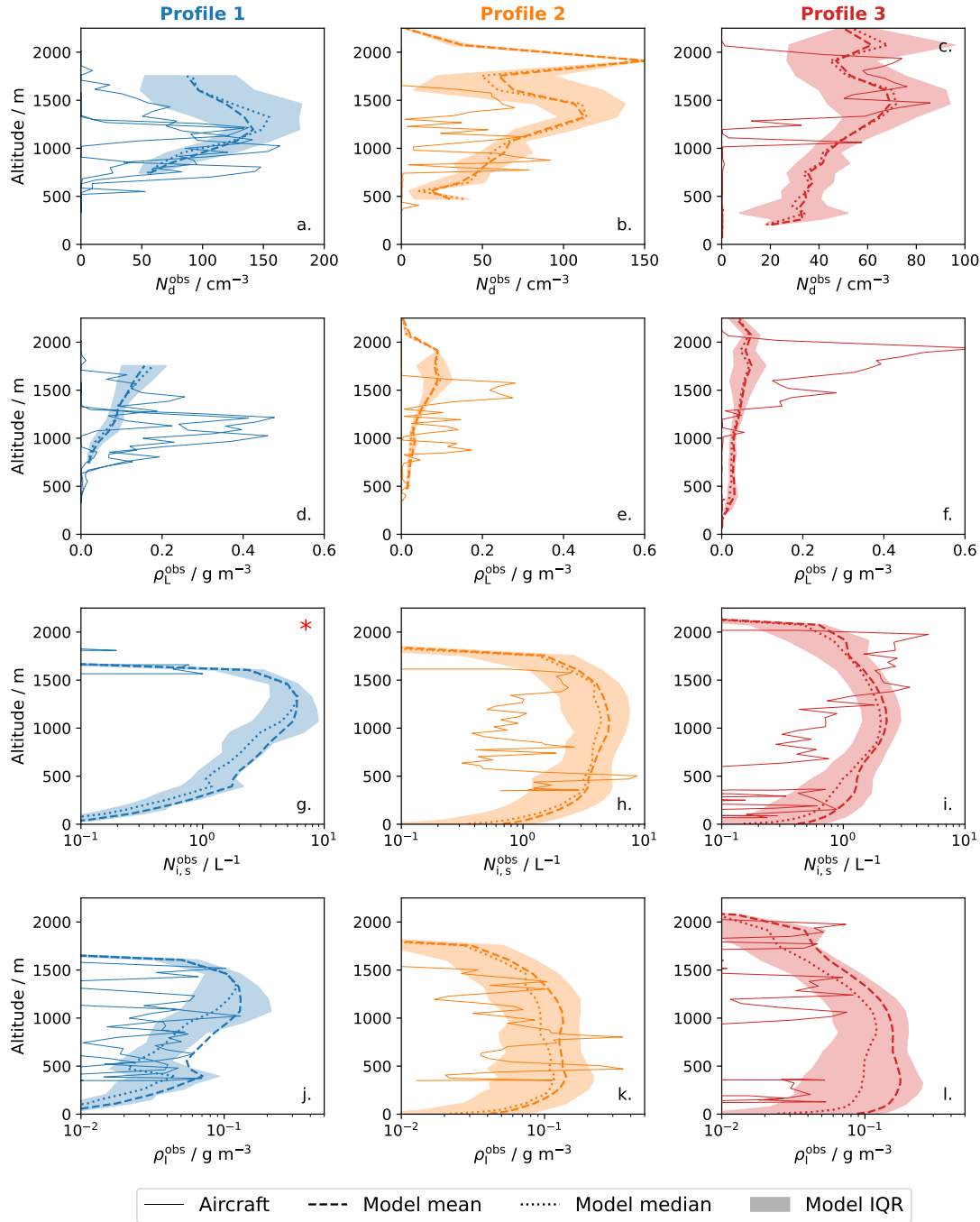


Figure 4.5: Profiles of hydrometeor measurements made by the FAAM aircraft compared to the model. Each column represents one of three profiles, with colours corresponding to those in Figure 4.4. From top to bottom, measurements along each row are: cloud droplet number concentration (N_d^{obs}), measured by the CDP; liquid water content (ρ_L^{obs}), measured by the Nevzorov hot-wire probe; snow concentration ($N_{i,s}^{obs}$), measured by the two-dimensional stereo probe (2-DS); ice water content (ρ_I^{obs}), also measured by the Nevzorov. Aircraft measurements are averaged over 10 s, which is approximately the time taken for the aircraft to traverse a grid box. These averages include measurements of zero. Shaded areas denote the interquartile range (IQR) of model data. Note that x-scales differ for panels (a), (b) and (c). Panel (g) is marked with an asterisk to denote that 2-DS data was largely unavailable for this profile.

Quantity	Profile	Model ($\mathcal{M}$)	Instrument ($\mathcal{I}$)	$\mathcal{M} - \mathcal{I}$	$\mathcal{M}/\mathcal{I}$
$N_d^{\text{obs}} / \text{cm}^{-3}$	1	111.1	93.4	17.7	1.19
	2	80.1	60.3	19.8	1.32
	3	55.4	61.3	-5.87	0.90
$\rho_L^{\text{obs}} / \text{g kg}^{-1}$	1	0.070	0.189	-0.119	0.36
	2	0.057	0.139	-0.082	0.41
	3	0.049	0.261	-0.222	0.19
$N_{i,s}^{\text{obs}} / \text{L}^{-1}$	1	5.85	-	-	-
	2	3.56	1.18	1.76	2.70
	3	0.58	1.48	-0.89	1.98
$\rho_I^{\text{obs}} / \text{g kg}^{-1}$	1	0.079	0.042	0.038	1.90
	2	0.110	0.085	0.024	1.29
	3	0.132	0.045	0.087	2.96
$\rho_T^{\text{obs}} / \text{g kg}^{-1}$	1	0.149	0.231	-0.082	0.65
	2	0.167	0.224	-0.057	0.75
	3	0.182	0.306	-0.124	0.59

Table 4.2: Mean values of observable droplet number (N_d^{obs}), liquid water content (ρ_L^{obs}), ice and snow concentration ($N_{i,s}^{\text{obs}}$), ice water content (ρ_I^{obs}) and total water content ($\rho_T^{\text{obs}} = \rho_L^{\text{obs}} + \rho_I^{\text{obs}}$) in the three aircraft profiles compared to model values. 2-DS data was unavailable for the first profile.

and ice water contents were measured by the Nevzorov probe (Korolev et al., 1998; Abel et al., 2014). Both are further discussed in Section 2.2.4. The concentration of ice particles and snow aggregates was measured by the two-dimensional stereo (2-DS) underwing probe (Stratton Park Engineering Company). The 2-DS measures hydrometeors with diameters between $15 \mu\text{m}$ and $1280 \mu\text{m}$ by capturing the shadow a passing particle creates when interrupting one of two crossing laser beams (Lawson et al., 2006). Particles are then categorised based on their irregularity O , which is proportional to the ratio of their perimeter and area (Crosier et al., 2011). Highly irregular particles, which have $O > 1.4$, are thought to be ice-phase, while those with $1.2 < O < 1.4$ may be liquid or ice-phase (Lloyd et al., 2020). In this study, we choose highly irregular particles to represent snow particles, representing a lower bound on snow concentration.

To enable direct comparison between model and observations, the model particle size distributions of liquid droplets, ice and snow were calculated using Eq. 3.1. Model concentrations were then obtained by integrating these distributions over the range of diameters observable by the CDP and 2-DS respectively. This gave N_d^{obs} , the observable cloud droplet concentration, and $N_{i,s}^{\text{obs}}$, the observable ice and snow concentration. To obtain the observable liquid water content, ρ_L^{obs} , grid cells with liquid cloud fractions lower than 0.1 were discarded, ensuring that cloud-free air in the model was not compared to measurements made in cloud. Similarly, to obtain ice water content, ρ_I^{obs} , grid cells with ice cloud fraction lower than 0.1 were discarded. The same constraint was applied when calculating N_d^{obs} and $N_{i,s}^{\text{obs}}$.

Compared to CDP (Figure 4.5a, b and c) and 2-DS measurements (Figure 4.5g, h and i), the

model captures N_d^{obs} and $N_{i,s}^{\text{obs}}$ reasonably well, though $N_{i,s}^{\text{obs}}$ is overestimated. Comparing cloud-average N_d^{obs} (Table 4.2), there is an inconsistent bias in N_d^{obs} between the three profiles. During Profiles 1 and 2, the model overestimated N_d^{obs} by 18 cm^{-3} and 20 cm^{-3} respectively. In the case of Profile 2, this was an overestimate by 33 %, the worst relative difference of the three profiles. In contrast to the first two profiles, the model underestimated N_d^{obs} during Profile 3 by 6 cm^{-3} .

Though the magnitude of differences in the measured and modelled $N_{i,s}^{\text{obs}}$ are larger than those of N_d^{obs} , the magnitude of the differences in $N_{i,s}^{\text{obs}}$ is within the uncertainty on the INP measurements which underpin the production of ice and snow. The overestimate of $N_{i,s}^{\text{obs}}$ during Profile 3 is consistent with the underestimate of N_d^{obs} during Profile 3. This may be linked to grid-box temperatures during Profile 3 being $(0.7 \pm 0.4) \text{ K}$ lower than those measured by the aircraft (shown in Figure C.2).

The model was also biased cold by $(0.5 \pm 0.6) \text{ K}$ during Profile 1 and $(0.6 \pm 0.4) \text{ K}$ during Profile 2. This may go some way to explaining the significant underestimate of ρ_L^{obs} which is 2.4 to 5.3 times too low across the three profiles (Figure 4.5d, e and f), since a 1 K decrease in temperature results in a 20 % to 80 % increase in INP concentrations, which control the ice particle concentration here given cloud is outside the Hallett-Mossop temperature range. Similarly, ρ_I^{obs} is generally overestimated, being 1.3 to 3 times the instrument value. The overproduction of ice in the model is consistent with previous CASIM work using aerosol-unaware ice nucleation by Huang et al. (2025) and Smith et al. (2025) (described in Section 3.3). Since the N_d^{obs} is well-represented in these profiles, this suggests droplets are too small, which may be indicative of a too-strong Wegener-Bergeron process. Despite the over-prediction of ρ_I^{obs} , the total water content (ρ_T^{obs}) was consistently underestimated by 40 % to 50 %.

The model shows varying degrees of ability at capturing the vertical structure of the clouds. Peak N_d^{obs} occurs at approximately coincident heights in each of the profiles, though in Profile 1 and 2, N_d^{obs} is overestimated higher in the cloud and underestimated lower in the cloud. In Profile 3, the vertical profile of $N_{i,s}^{\text{obs}}$ is well-matched to the measurements in the cloud, though in the precipitation below the cloud, the $N_{i,s}^{\text{obs}}$ is underestimated by up to an order of magnitude. However, the overestimation of ρ_I^{obs} by a lower degree at this altitude occurs since the ice particle size distribution in the model is biased toward lower sizes than those in the detection range of the 2-DS.

4.4.2 Removal of INPs

To quantify the change in INP concentration through the CAO, average profiles of INP concentrations and INP-relevant quantities were calculated in six areas of approximately 50 km^2 along the direction of the prevailing wind. These areas are shown in Figure 4.3a, with colours that will henceforth be used consistently. Profiles of the liquid and ice concentrations along the trajectory are shown in Figure 4.6. Box 1 (purple) is farthest upstream and is over

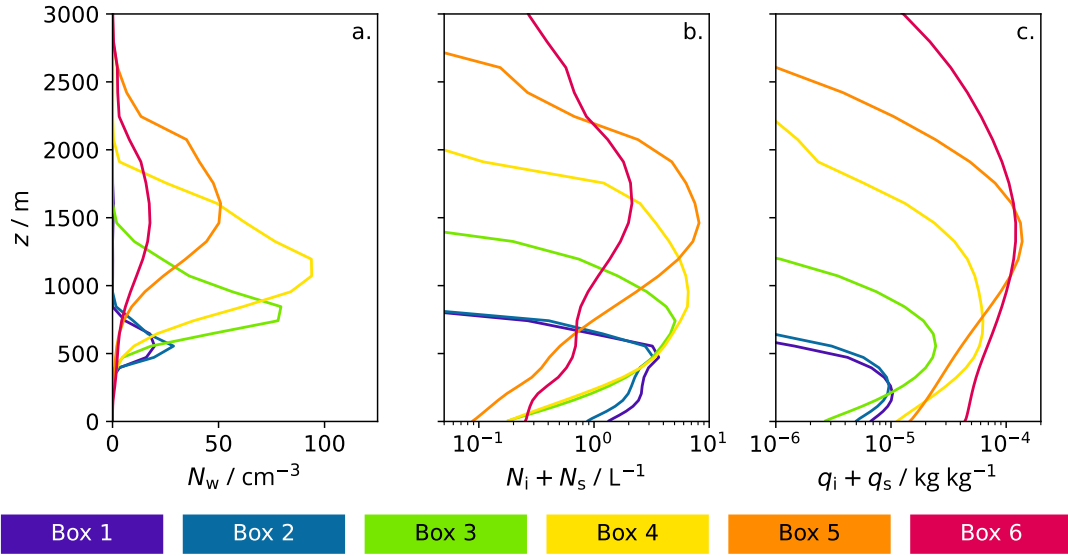


Figure 4.6: Profiles of grid-box mean (a) N_w , (b) combined N_i and N_s and (c) combined q_l and q_s along the trajectory of the CAO. Values are averages between 11:00 and 12:00 UTC on 29 March 2022. Line colours correspond to average profiles over areas indicated by the boxes in Figure 4.3a, which follow a rainbow pattern from purple upstream to red downstream.

largely closed pack ice with small amount of thin mixed-phase cloud at approximately 500 m in altitude. Box 2 (blue) is predominantly over the marginal ice zone where some stratus cloud is initiated, while box 3 (green) is largely ice-free and sees early-stage stratocumulus cloud streets across the width of the box. Box 4 (yellow) sees developed stratocumulus cloud streets and covers most of the measurements discussed in Section 4.4.1, while box 5 (orange) sees significant deepening of the cloud as the SCT occurs. Finally, box 6 (red) is farthest downstream and contains open cumulus cells.

Figure 4.7 shows average profiles of INP concentrations and insoluble aerosol through each of these boxes. INP concentration is strongly temperature-dependent, so to measure the removal of INPs, we consider INP at a reference temperature as this is proportional to the concentration of insoluble aerosol (Eq. 4.1). Since all clouds from boxes 1 to 6 access temperatures below -15°C , -15°C was chosen as the reference temperature for INP concentration as INPs in the clouds which were active at this temperature were always able to initiate freezing. Figure 4.7b shows that the below-cloud INP concentration reduces downstream – the average below-cloud INP concentration in box 2, where CAO clouds first initiate, is $(1.12 \pm 0.06) \text{ L}^{-1}$, while for box 6 it is $(0.644 \pm 0.005) \text{ L}^{-1}$. As shown in Figure 4.7c, the reduction in below-cloud INP concentration is proportional to the reduction in below-cloud total insoluble aerosol concentration (i.e. both contained in and outside of hydrometeors), N_{insol} , which is from $(14 \pm 1) \text{ cm}^{-3}$ to $(8.0 \pm 0.1) \text{ cm}^{-3}$.

To compare the reduction in INP concentrations to measurements, we define the drop-off rate ψ , such

$$\psi(T) = \frac{N_{\text{upstream INP}}(T)/N_{\text{downstream INP}}(T)}{C}, \quad (4.3)$$

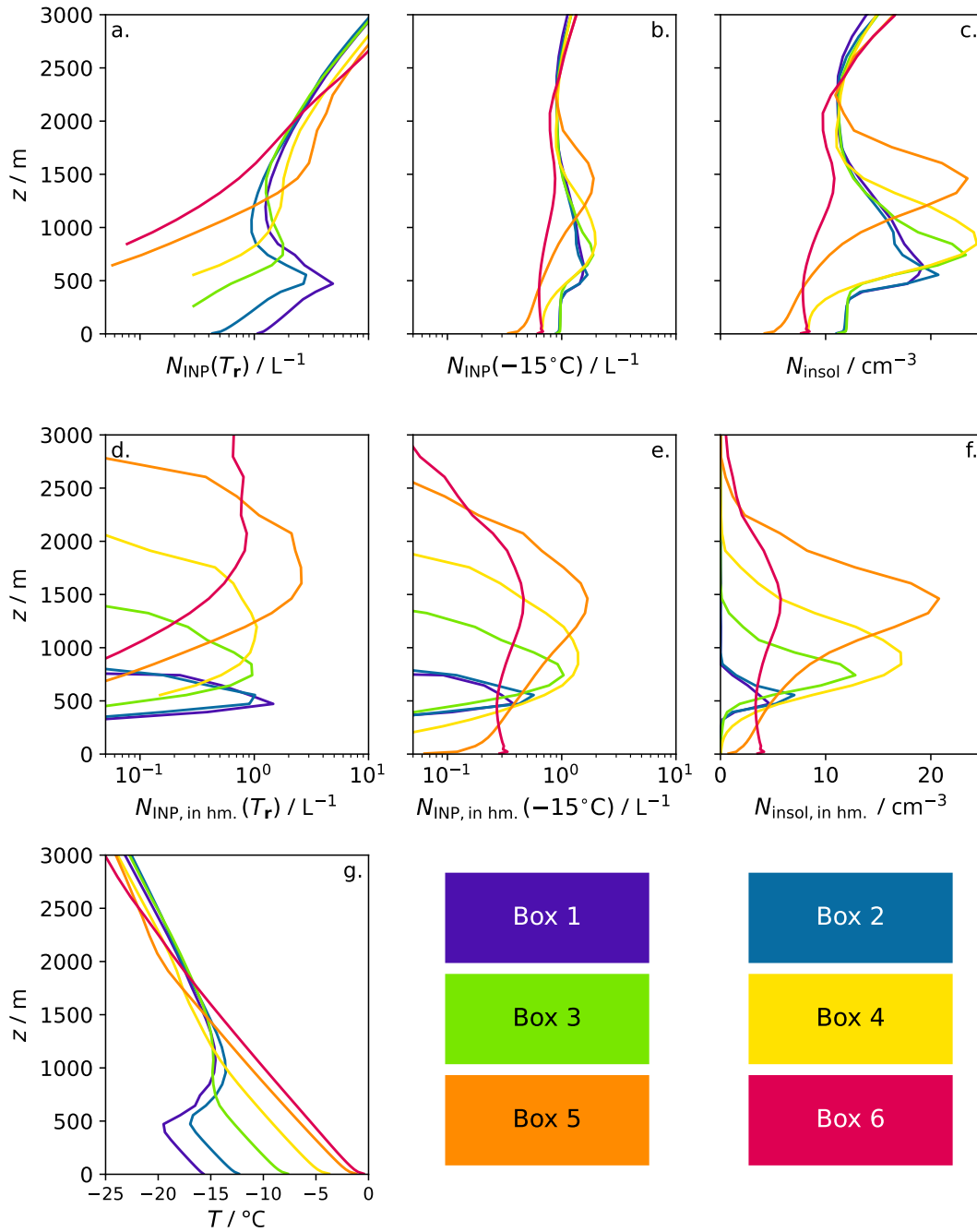


Figure 4.7: Profiles of INP and insoluble aerosol concentrations along the trajectory of the CAO. Line colours correspond to average profiles over areas indicated by the boxes in Figure 4.3a, which follow a rainbow pattern from purple upstream to red downstream. Panels (a) and (b) show profiles of the INP concentration, n_{INP} at the local temperature and at -15°C . The total number concentration of insoluble aerosol, N_{insol} , is shown on panel (c). The number of INPs within hydrometeors, $n_{\text{INP, in hm.}}$, is shown at local temperature and at -15°C in panels (d) and (e), respectively, while the number of insoluble aerosol particles in hydrometeors is shown in panel (f). To assist with the interpretation of the temperature-dependent concentrations shown in panels (a) and (d), the temperature profile is provided in panel (g). All values are averages between 11:00 and 12:00 UTC on 29 March 2022.

Comparison locations	$\psi_{\text{obs}}(-15^\circ\text{C}) / \text{km}^{-1}$	$\psi_{\text{model}}(-15^\circ\text{C}) / \text{km}^{-1}$	$\frac{\psi_{\text{obs}}(-15^\circ\text{C})}{\psi_{\text{model}}(-15^\circ\text{C})}$
Below Sc filter to below Cu filter	0.011 ± 0.005	0.0042 ± 0.0011	2.5
Below Sc filter to Andenes	0.23 ± 0.14	0.0019 ± 0.0003	120

Table 4.3: Comparison of the INP concentration drop-off factor between observations made both on the aircraft and at Andenes, $\psi_{\text{obs}}(-15^\circ\text{C})$, and in the model, $\psi_{\text{model}}(-15^\circ\text{C})$. ψ is defined by Eq. 4.3.

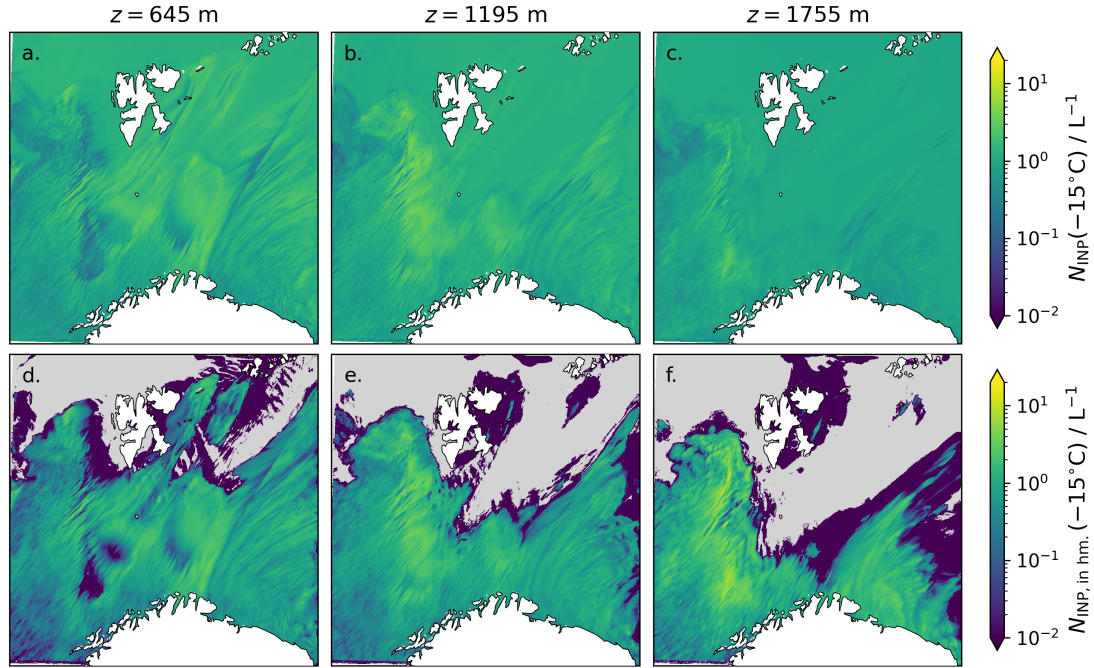


Figure 4.8: Average INP concentrations at -15°C between 11:00 and 12:00 UTC at three cloud-relevant heights. Panels (a), (b) and (c) show $n_{\text{INP}}(-15^\circ\text{C})$ at heights $z = 645 \text{ m}$, 1195 m and 1755 m respectively, while the concentration of INPs contained within hydrometeors, $n_{\text{INP, hm.}}(-15^\circ\text{C})$, is shown in panels (d), (e) and (f). All values are averages between 11:00 and 12:00 UTC on 29 March 2022.

where C is the distance between the upstream and downstream locations. Larger values of ψ indicate a faster rate of reduction in INP concentration. Values of $\psi(-15^\circ\text{C})$ are shown in Table 4.3. The model under-predicts the reduction in INP concentration between the below-cloud filter samples by a factor of 2.5 relative to the observations, though the values of $\psi_{\text{obs}}(-15^\circ\text{C})$ and $\psi_{\text{model}}(-15^\circ\text{C})$ agree within 1.3σ . In contrast, the model dramatically under-predicted the reduction in INP concentration between the below Sc aircraft filter sample and Andenes, with $\psi_{\text{model}}(-15^\circ\text{C})$ being 120 times smaller than $\psi_{\text{obs}}(-15^\circ\text{C})$.

Although the model shows a reduction in below-cloud INP concentrations downstream, the three-dimensional distribution of INP throughout the cloud system is complex, as shown in Figure 4.8, which shows maps of $N_{\text{INP}}(-15^\circ\text{C})$ and $N_{\text{INP, in hm.}}(-15^\circ\text{C})$, the number concentration of INPs contained within hydrometeors. These quantities are shown at three reference model levels, $z_1 = 645 \text{ m}$, $z_2 = 1195 \text{ m}$ and $z_3 = 1755 \text{ m}$. z_1 is approximately

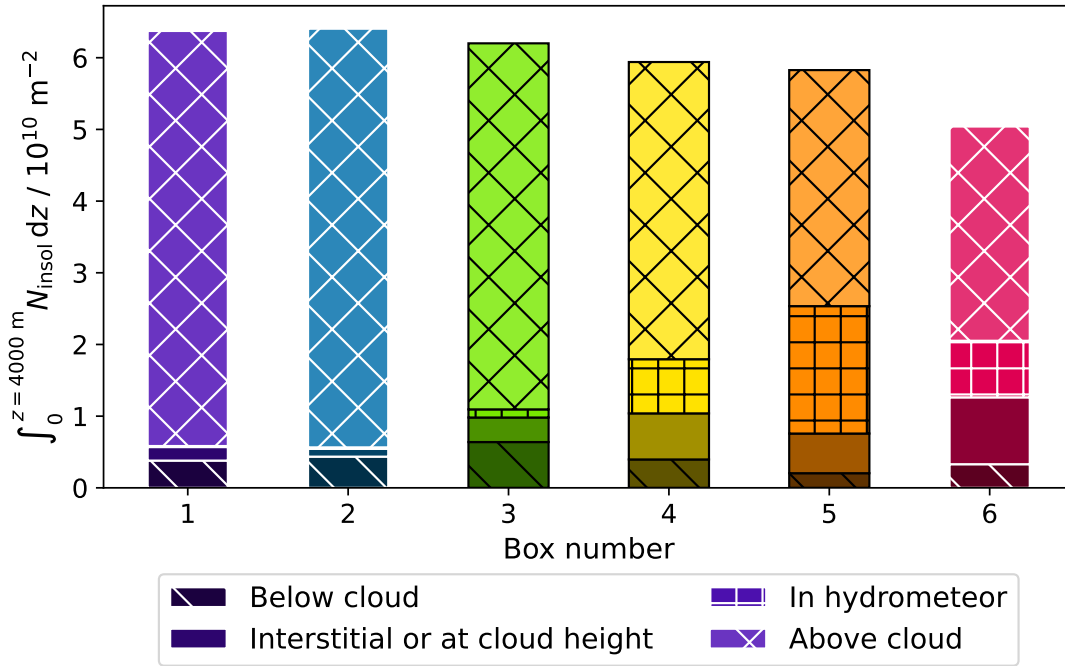


Figure 4.9: Column-integrated insoluble aerosol concentrations in each analysis box, averaged between 11:00 and 12:00 UTC on 29 March 2022.

representative of the cloud base in the south-west quadrant of the domain, though it is slightly below it in places. z_2 is approximately representative of cloud-top in box 3, and of the cloud bulk in boxes 4 to 6. z_3 is approximately representative of cloud-top in boxes 4 to 5, and of the cloud bulk in box 6.

The observed below-cloud reduction in INP concentrations is visible in Figure 4.8a, which shows $N_{\text{INP}}(-15^\circ\text{C})$ at z_1 . At z_2 , more INPs are ingested into the cloud through entrainment at cloud-top. This is visible in Figure 4.8b and Figure 4.8e, where $N_{\text{INP}}(-15^\circ\text{C}) \approx N_{\text{INP, in hm.}}(-15^\circ\text{C})$ immediately downstream of Bjørnøya, and is greater than at z_1 . The same effect is visible in further downstream in Figure 4.8c and Figure 4.8f, which show that $N_{\text{INP, in hm.}}(-15^\circ\text{C})$ at z_3 is greater than $N_{\text{INP, in hm.}}(-15^\circ\text{C})$ at z_2 and z_1 in this region.

The competition between the ingestion of INPs from above cloud and the removal of INPs results in remarkably consistent peak values of $N_{\text{INP}}(-15^\circ\text{C})$ in profiles of the cloud (Figure 4.7b). Indeed, the peak values of $N_{\text{INP}}(-15^\circ\text{C})$ in the INP profiles are remarkably consistent between all six analysis boxes, with a range of 1.4 L^{-1} to 1.9 L^{-1} .

To assess the net effect of the competition between the entrainment of above-cloud aerosol and removal of aerosol, Figure 4.9 shows column integrals of N_{insol} from 0 m to 4000 m, averaged spatially within boxes and temporally between 11:00 UTC and 12:00 UTC. Integrals were calculated above-, below- and in-cloud, and in hydrometeors over the full column. Columns without cloud were excluded from the calculation. The upper limit of 4000 m was chosen to be sufficiently above cloud top to avoid undercounting in-cloud insoluble aer-

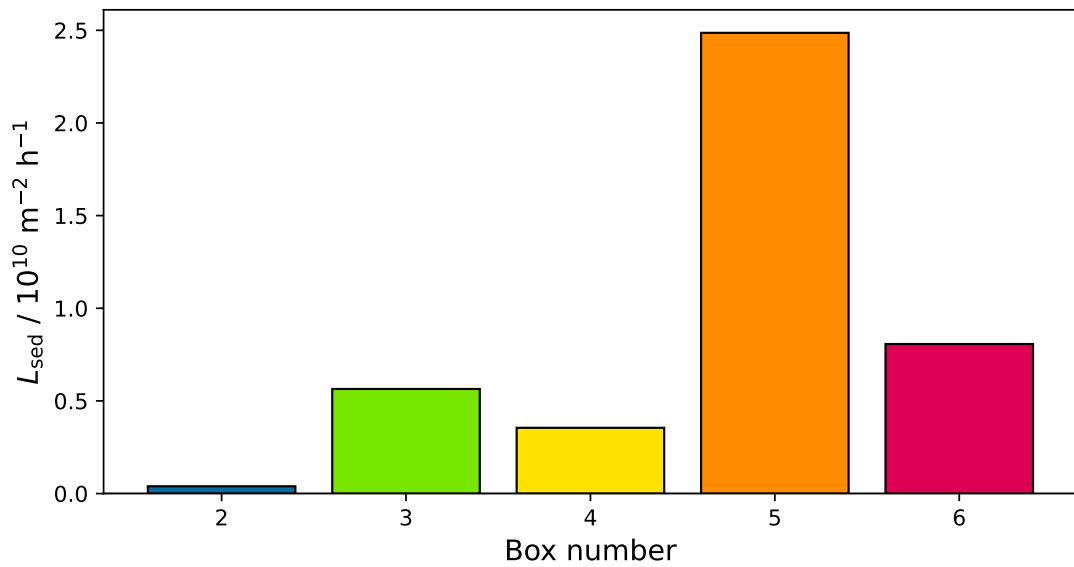


Figure 4.10: Column-integrated loss of insoluble aerosol via the sedimentation process in each of the analysis boxes, L_{sed} . The flux is integrated over the column between 0 m and 4000 m and averaged between 11:00 and 12:00. Rates are of similar magnitude in the hour prior.

osol, but low enough to avoid insoluble aerosol high in the free troposphere that could not interact with the cloud. By this time, aerosol concentrations in each of the columns had reached an approximately steady state and the lateral flux divergence in each column was negligible relative to the column totals. Additionally, dry deposition fluxes were less than $1 \times 10^6 \text{ m}^{-2} \text{ h}^{-1}$. Consequently, changes in column totals can act as a proxy for vertical flux divergences. The total below-cloud insoluble aerosol does not consistently reduce between the boxes, though this is due to the increased cloud base height. As the cloud deepens between boxes 3 and 5, the total interstitial and in-hydrometeor insoluble aerosol concentration increases by depleting the above-cloud aerosol through entrainment. This entrainment of aerosol did not fully compensate for the removal of INPs, as the column total insoluble aerosol steadily reduced across each of the profile boxes.

The INP removal was driven by precipitation. Figure 4.10 shows L_{sed} , the flux of insoluble aerosol lost due to precipitation, which occurs via the model's sedimentation process (see Table 3.1 and Table 3.2). This was by the dominant sink of INPs, with the magnitude of L_{sed} in each box correlating strongly with the reduction of column-integrated INP concentration between boxes shown in Figure 4.9. This is particularly notable in box 6, which featured the highest precipitation rates of the domain, and where the total in-hydrometeor aerosol decreased, significantly outcompeting the entrainment of aerosol into the column. However, the removal was not especially efficient over the domain, with the total column insoluble aerosol concentration being reduced by approximately 15 % between box 1 and box 6.

While this aerosol-aware modelling did show that INP concentrations reduced downstream due to the removal of aerosol through precipitation, the rate of reduction in INP concentra-

tions was 2.5 times smaller than observed by the aircraft. However, the lack of repeated INP measurements makes it difficult to judge how statistically significant an underestimate this is. A possible reason for the lower removal rate in the model may be the overproduction of ice relative to observations, resulting in smaller ice and snow sizes and thus reduced precipitation. An alternative explanation may be the use of an INP parameterisation based on aerosol number concentration rather than aerosol surface area. Despite the physical links between aerosol surface area and ice-nucleating activity (e.g. Niemand et al., 2012; Vali et al., 2015; Kanji et al., 2017; Vali, 2019), aerosol number was chosen as the property linking aerosol and ice nucleation in this study. This choice was made since the values of individual measurements of $N_{\text{INP}}/N_{\text{aer}}$ upstream had less spread than those of $N_{\text{INP}}/S_{\text{aer}}$, reducing uncertainty on the parameterisation. However, assuming sites are uniformly distributed across the size distribution of insoluble aerosol, larger aerosols contribute a greater proportion of the surface area and thus are more likely to nucleate ice. Removal of these aerosols through ice processes would proportionately reduce the total insoluble aerosol surface area more strongly than the total insoluble aerosol number, resulting in lower INP concentrations at the next model timestep. Consequently, a surface area-based INP parameterisation may demonstrate more efficient removal of INPs.

These hypotheses might account for the discrepancy in INP concentration reduction rate between the aircraft measurements of INP concentration. However, it seems unlikely these hypotheses alone could account for the extremely large discrepancy in the observed and modelled reduction in INP concentration between the aircraft measurement and Andenes, which differed by a factor of 120. Instead, it seems likely that some of this discrepancy arises due to differences between the techniques used to measure INPs on the aircraft and at Andenes. Indeed, discrepancies between techniques means that differences in INP concentration of one order of magnitude are not uncommon when comparing measurements of the same INP population using well-characterised methodologies (Hiranuma et al., 2019; Lacher et al., 2024). While the aircraft measured INP concentrations using the drop-on technique described in Section 2.2.2, the measurements at Andenes used an impinger technique based on the DRoplet Ice Nuclei Counter Zurich (DRINCZ) (David et al., 2019; Gjelsvik et al., 2025). When measuring samples of NX-illite, a mineral dust, the DRINCZ wash-off technique compared well to measurements of mineral dust made by Sanchez-Marroquin et al. (2021) using a wash-off technique. However, this wash-off technique has undercounted INPs relative to the drop-on technique, particularly when analysing samples of INPs with biological components (Sanchez-Marroquin et al., 2021; Daily et al., 2025; Robinson et al., 2026). Given that it was hypothesised that the dominant INPs during ACAO were fertile soil dusts, this suggests that removal was not as strong as comparison between the aircraft and Andenes measurements would suggest. It would be advantageous in future field measurements for different locations to use the same technique to minimise such biases.

4.4.3 INP concentrations at cloud temperature

As described above, $N_{\text{INP}}(-15^\circ\text{C})$ is a useful quantity to compare observed INP concentrations and assess removal of INP. However, the steep dependence of INP on temperature means that the INP concentration can vary strongly across a domain, even if the aerosol concentration is identical. Even where it is not, the temperature dependence of ice nucleation often dominates the dependence on aerosol concentration. For instance, according to Eq. 4.1, a cooling of 2°C from -10°C to -12°C would increase the INP concentration of an aerosol sample more than doubling the insoluble aerosol concentration. In this case study, such a change in temperature equates to an increase in height of approximately 200 m to 300 m, less than the height of the stratocumulus cloud and much less than the height of cumulus cloud. As such, the INP concentration that is relevant for a cloud at position $\mathbf{r}$ is $N_{\text{INP}}(T(\mathbf{r}))$.

Vertical profiles of $N_{\text{INP}}(T_{\mathbf{r}})$ are shown in Figure 4.7a, alongside $N_{\text{INP, in hm.}}(T_{\mathbf{r}})$, the number concentration of INP in hydrometeors at position $\mathbf{r}$, in Figure 4.7d and the temperature in Figure 4.7g. The below-cloud temperatures increase from a minimum of $T = -20^\circ\text{C}$ in box 1 to $T > -5^\circ\text{C}$ in box 6, and Figure 4.7a shows that there is a corresponding decrease in $N_{\text{INP}}(T_{\mathbf{r}})$ for $z < 500$ m from box 1 to box 4, before temperatures become higher than those where any freezing was observed. However, boxes 1 and 2 experience temperature inversions at approximately 500 m and 850 m respectively, while the lapse rate in boxes 3, 4 and 5 decreases above 1000 m. This means that at $z = 2600$ m box 6 is approximately 4°C cooler than box 1 and $N_{\text{INP}}(T_{\mathbf{r}})$ is 9 times larger. As such, the latitudinal surface temperature gradient (which warms towards the south) and the removal of INPs is counteracted by the vertical temperature gradient experienced by the deepening cloud as the CAO develops.

This means that peak values of $N_{\text{INP, in hm.}}(T_{\mathbf{r}})$, shown in Figure 4.7d, are similar throughout the CAO as the deepening cloud activates more INP sites at lower temperatures. Indeed, in boxes 2 to 4, which represent the initiation of the CAO cloud streets to the developed Sc, the maximum values of the profile of $N_{\text{INP, in hm.}}(T_{\mathbf{r}})$ vary by an extremely small amount, and are between 0.96 L^{-1} and 1.05 L^{-1} . Of the six boxes, box 5 has the highest values of $N_{\text{INP, in hm.}}(T_{\mathbf{r}})$; these occur as box 5 experiences the lowest temperatures of any of the clouds at an altitude which is well inside the limits of the cloud.

In Section 2.4, we discussed the apparent contradiction between the measurements made during ACAO, which showed a reduction in INP concentrations downstream, and a remote sensing study of CAOs in the same region by Murray-Watson and Gryspeerdt (2024). In their study, they observed increased ice concentrations downwind in CAOs and consequently inferred increased INP concentrations downstream, attributing this increase to sea-spray aerosol.

Figure 4.6c shows that ice mixing ratio also increased downstream in our model, despite concentrations of INPs being reduced when measured at a reference temperature. Our

study did not include an INP parameterisation for sea-spray aerosol, but given the significantly lower ice-nucleating ability of sea-spray aerosol compared to fertile dusts, it seems unlikely that sea-spray would have contributed significantly to the INP population at temperatures important for these clouds.

Instead, these two perspectives can be reconciled by considering INP concentrations at the temperature they are active rather than relative to the aerosol concentration. Downstream, taller CAO clouds access temperatures which are lower than shallower clouds farther upstream and so INP concentrations may increase without any change to the aerosol concentration. This also implies that it is difficult to make inferences about INP concentrations from analysis of ice alone in the way that droplet concentrations can be used to make inferences about CCN concentrations. While an increase in INPs implies an increase in ice concentrations, an increase in ice concentration does not necessarily imply an increase in INP concentration, even when SIP is absent.

4.5 Conclusions

In this study, we performed the first interactive, aerosol-aware simulation of INPs in a CAO, based on measurements made during the ACAO campaign. The model was largely able to capture the properties of the cold-air outbreak clouds, though as with other studies using aerosol-unaware implementations of CASIM, ice-phase hydrometeor number concentrations were too high. As in the campaign, a reduction in the number of INPs was simulated downstream when considering INP concentration at a reference temperature, though the rate of reduction in INP concentration was 2.5 times smaller than that which was observed. However, with just two measurements, it is difficult to assess how meaningful this underestimate is. The model was unable to account for the difference in concentration between airborne INP measurements and hilltop INP measurements further downwind at Andenes, suggesting differences in the sensitivities of the two techniques employed. Future measurements aiming to understand the transport of INPs should aim to use consistent techniques along the entire cloud system.

In the absence of substantial removal of insoluble aerosol, the changes in cloud temperature controlled the concentration of INPs. Deepening clouds farther downstream reached lower temperatures and thus were able to maintain a steady INP concentration throughout the CAO. This consistent peak INP concentration, along with the limited removal of INPs in the model despite full aerosol processing, suggests that for CAOs with similar temperature profiles, aerosol-aware INP parameterisations may add unnecessary computational complexity to the model relative to passive ice nucleation. Aerosol-unaware parameterisations may be sufficient to model INP concentrations in CAOs within the uncertainty provided by regional in-situ measurements, provided they are based on measurements sensitive enough to capture the full range of INP concentrations. However, this conclusion may be an artefact of the particular latitudinal temperature gradient and INP concentrations in this case study.

Further work exploring CAOs in high- and low-INP concentration environments, with strong and weak temperature gradients through the outbreak could establish if an aerosol-aware parameterisation is necessary.

While INPs were not strongly removed in the model, it is possible that the removal of INPs may have been suppressed by a deficiency in the way in which INPs are represented. This is explored in Chapter 5.

Chapter 5

Tracking active sites matters when modelling the INP removal rate in a cold-air outbreak

Abstract

When clouds evolve, INPs are removed by precipitation. However, models often make the simplifying assumption that there is a constant relationship between the aerosol concentration and the number of INPs, resulting in an overestimate of INP concentration. To test the magnitude of this bias, we implemented a new INP-limited ice-nucleation scheme in the Met Office Unified Model which tracks a population of INPs which can be removed. By using this new scheme, we were able to simulate the strong reduction in INP concentrations observed through the evolution of a cold-air outbreak which could not be represented by a traditional ice nucleation scheme. However, we were unable to simultaneously represent the magnitude of the INP concentration and the rate of INP removal and so we suggest a new method of simulating INPs to reconcile these.

5.1 Introduction

In Chapter 4, we presented model simulations of a case study of a CAO from the ACAA campaign where observations suggested that INPs were removed by precipitation. These simulations showed that INPs were weakly removed as the CAO developed, but that the removal was compensated for by entrainment of INPs at cloud-top. As such, the temperature controlled the INP concentration, which remained steady throughout the outbreak.

To perform this modelling, we used an INP parameterisation which assumed the singular hypothesis of ice nucleation. To perfectly represent the singular hypothesis, a model would

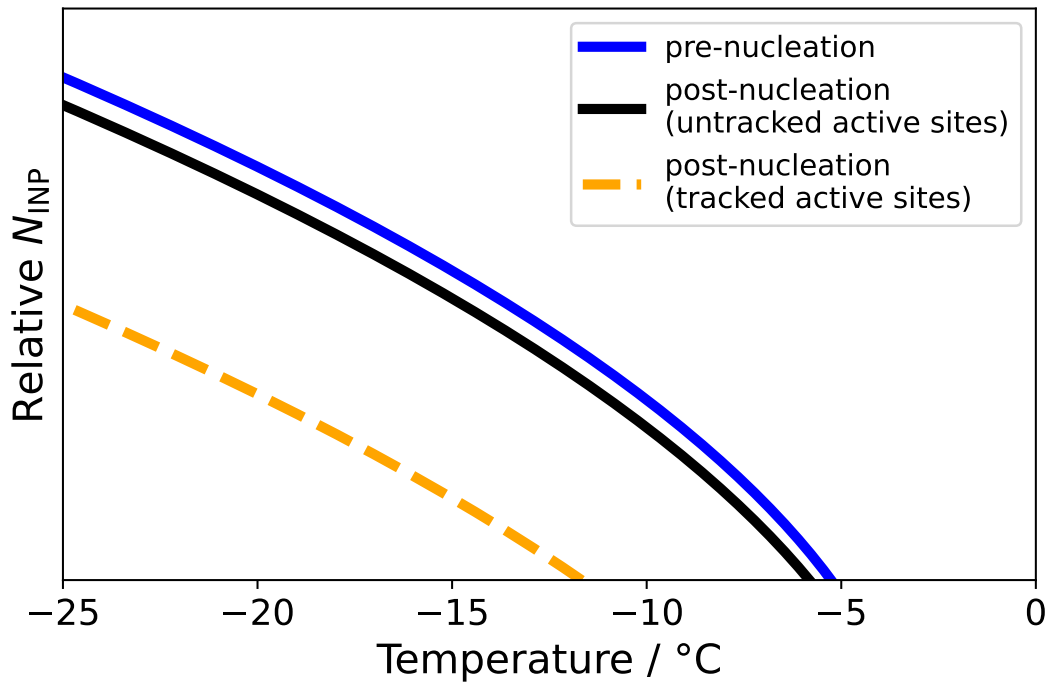


Figure 5.1: Schematic showing comparing INP spectra after ice nucleation in when active sites are or are not tracked.

need to trace each INP according to its characteristic ice nucleation temperature (Knopf and Alpert, 2023). This is computationally impossible, so most aerosol-aware ice nucleation schemes make the simplifying assumption that there is a constant relationship between the amount of aerosol and the INP concentration. In this assumption, active sites for ice nucleation are not tracked through the simulation.

5.1.1 Untracked active sites

The INP parameterisation used in Chapter 4 was akin to the DeMott et al. (2010) parameterisation, which prognoses the INP concentration as a fraction of the insoluble aerosol concentration. As discussed in Section 1.2.5, this, like other aerosol-aware bulk microphysical representations of INPs which use the singular hypothesis, takes the form of

$$\frac{dN_i}{dt} = \frac{N_{\text{INP}}}{\Delta t} = \frac{f(T)g(\mathcal{A})}{\Delta t}, \quad (5.1)$$

where Δt is the model timestep and f is a function of temperature that is multiplied by g , a function of an aerosol property $\mathcal{A}$. This aerosol property, $\mathcal{A}$, is typically the number concentration or surface area of a subset of the aerosol particles, such as the mineral dust, but only considers aerosol particles which have not yet nucleated ice. After an ice nucleation event, $g(\mathcal{A})$ should reduce, since there are fewer INPs available. However, since active sites are not tracked, the form of the function g does not evolve after ice nucleation events, so the magnitude of $g(\mathcal{A})$ can only change if the value of $\mathcal{A}$ changes. Since INPs are rare relative

to the aerosol population ($f(T) \ll 1$), ice nucleation on INPs has little effect on the aerosol population:

$$\begin{aligned} g(\mathcal{A}(t + \Delta t)) &= g(\mathcal{A}(t)) - g(N_{\text{INP}}) \\ g(\mathcal{A}(t + \Delta t)) &= g(\mathcal{A}(t)) - g(f(T)g(\mathcal{A})) \\ g(\mathcal{A}(t + \Delta t)) &\approx g(\mathcal{A}(t)). \end{aligned} \quad (5.2)$$

This means that between timesteps, $g(\mathcal{A})$ remains approximately constant and thus N_{INP} also remains approximately constant, as shown in Figure 5.1. Accordingly, the lack of evolution of g means that aerosol does not act as a particularly strong limit on ice nucleation. Instead, the aerosol population effectively acts as an infinite reservoir of INP since it is never efficiently reduced.

5.1.2 Tracked active sites

The misrepresentation of parameterisations which do not track active sites arises since $g(\mathcal{A})$ effectively assumes that ice nucleation sites are distributed uniformly and randomly across a large subset of aerosol. However, experimental evidence suggests that only a small fraction of the aerosol population carries an ice nucleation site, since $N_{\text{INP}} \ll N_{\text{aer}}$ (Vali, 2008; Herbert et al., 2014; Holden et al., 2019). Were INP calculated according to

$$\frac{dN_i}{dt} = \frac{N_{\text{INP}}}{\Delta t} = \frac{f(T)h(\mathcal{S})}{\Delta t}, \quad (5.3)$$

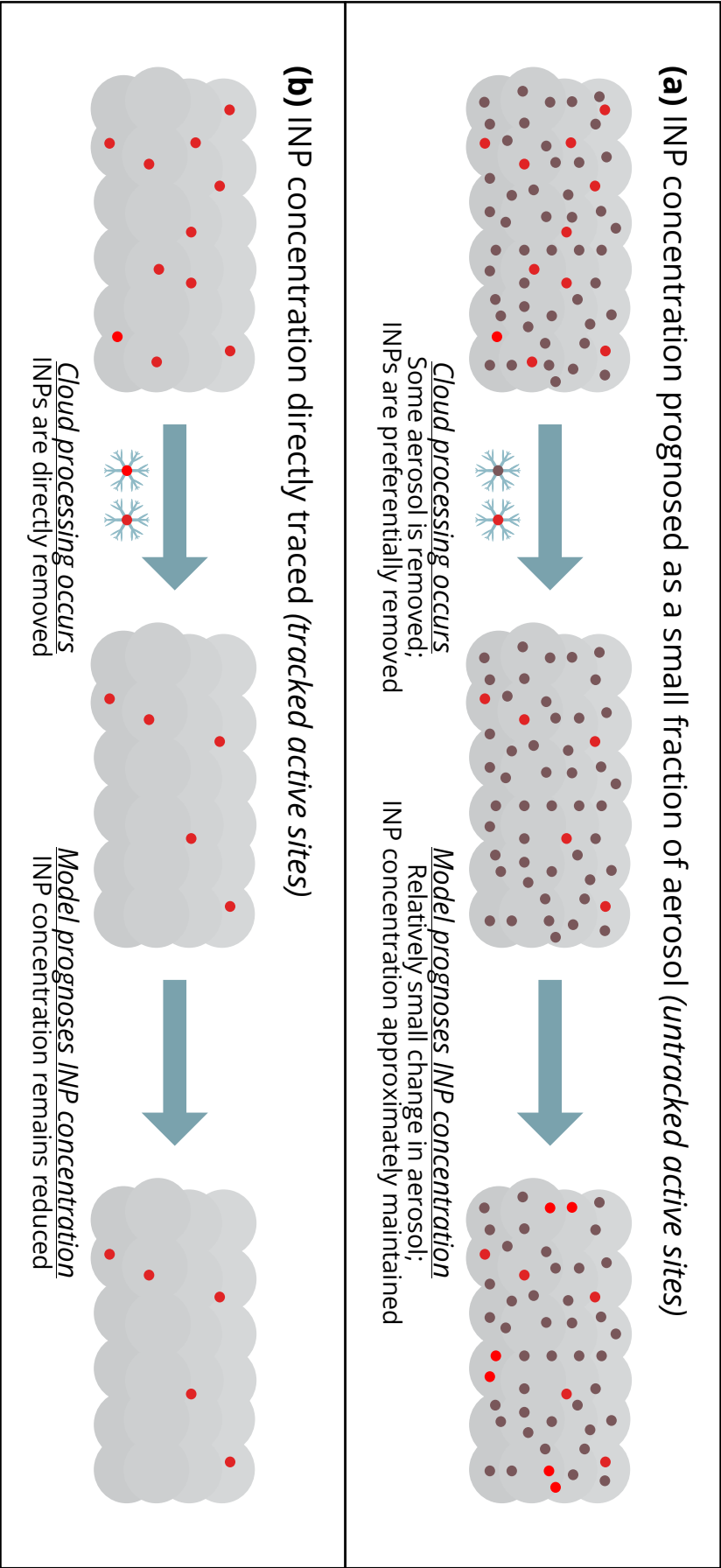
where h is a function of $\mathcal{S}$, the number of ice nucleation sites that are on INPs which have not yet frozen, we might expect that

$$\mathcal{S}(t + \Delta t) \ll \mathcal{S}(t), \quad (5.4)$$

if the majority of ice nucleation sites are contained on the INPs which were active and thus have frozen. This means that N_{INP} would be greatly reduced between timesteps, represented by the orange line in Figure 5.1.

5.1.3 Active site tracking and INP removal

The differences between these two alternative pictures of the distribution of ice nucleation sites across the aerosol population has consequences for modelling the removal of INPs from the cloud system, as shown in Figure 5.2a. When the freezing caused by INPs results in precipitation, INPs are preferentially removed from the cloud system compared to other aerosols, which may be washed out by scavenging. As such, the proportion of INPs which are removed should be much larger than the proportion of aerosols which are removed. However, since $g(\mathcal{A})$ remains constant, the two are linked. This means that in the absence of extremely strong removal of aerosol, the INP concentration will be approximately re-



Key: ● INP ● Aerosol which may nucleate ice *Ratio of INPs to aerosols exaggerated for clarity*

Figure 5.2: A schematic showing how different representations of INP can affect the modelling of INP removal in clouds.

Table 5.1: The simulations performed in this work.

Simulation	INP scheme	Magnitude of $\mathcal{P}$, relative to <i>Observed-15</i>
Default	As Section 4.3	<i>N/A, uses aerosol profile</i>
Abundant-15	INP-limited, $T_{\text{cap}} = -15\text{ }^{\circ}\text{C}$	10
Observed-15	INP-limited, $T_{\text{cap}} = -15\text{ }^{\circ}\text{C}$	1
Scarce-15	INP-limited, $T_{\text{cap}} = -15\text{ }^{\circ}\text{C}$	0.1
Abundant-20	INP-limited, $T_{\text{cap}} = -20\text{ }^{\circ}\text{C}$	36
Observed-20	INP-limited, $T_{\text{cap}} = -20\text{ }^{\circ}\text{C}$	3.6
Scarce-20	INP-limited, $T_{\text{cap}} = -20\text{ }^{\circ}\text{C}$	0.36

stored — effectively meaning that new INPs are artificially generated. In contrast, if INPs are tracked directly (Figure 5.2b), rather than as a function of the aerosol population, then an INP removed from the cloud system will stay removed (unless it is recirculated from an evaporated hydrometeor).

In Chapter 4, we showed that the model was unable to reproduce the rate of INP removal observed during a case study of a CAO. However, directly tracing INPs rather than prognosing their concentration from the aerosol concentration may allow us to reconcile model and measurements. In this chapter, we implement such a scheme, to ask whether INP removal can be better represented in a regional model by using an INP scheme which limits ice nucleation according to the number of INPs, and thus tracks active sites, rather than representing INPs as an unchanging fraction of a much larger aerosol population.

5.2 Model setup and hypotheses

To test the need for an alternative representation of INPs, we used the case study previously described in Section 4.2 with seven simulations using different model ice nucleation schemes and aerosol tracer values, summarised in Table 5.1.

The *default* simulation used the ice nucleation scheme described in Section 4.3, where INP concentration was diagnosed from the insoluble aerosol concentration, which was specified according to measurements of aerosol during the ACAO campaign.

We compared this to a new INP scheme, which we call *INP-limited*. In the *INP-limited* scheme, INPs are directly simulated rather than having their concentration calculated as a fraction of the insoluble aerosol. To achieve this without including a new model tracer, which would introduce significant computational complexity, the insoluble aerosol tracers in CASIM were repurposed to trace INPs. Using this, we can effectively simulate just the INP-active dust particles, neglecting the inactive dust particles.

In the *default* simulation, the insoluble aerosol tracer was initialised with a profile of insoluble aerosol concentration derived from measurements. Instead, in the *INP-limited* simu-

lation, the INP tracer was initialised with a profile of INP concentration, $\mathcal{P}$. Although the insoluble aerosol comprised a small fraction of the total aerosol concentration in the *default* simulation, the difference between the INP concentration and the insoluble aerosol concentration was added to the soluble aerosol to maintain the same total aerosol and thus CCN concentrations.

To derive $\mathcal{P}$, a reference temperature for ice nucleation, T_{cap} , was chosen. The average of the two upstream $N_{\text{INP}}(T_{\text{cap}})/N_{\text{aer}}$ measurements was then multiplied by the measured aerosol profile N_{aer} , making the assumption that $N_{\text{INP}}/N_{\text{aer}}$ was approximately consistent with height upstream.

We hypothesise that limiting the INP concentration will enhance the removal efficiency because an increasing fraction of the available INPs will form ice particles, be removed and not resupplied. This scheme allows us to explore increasingly INP-limited conditions by changing the magnitude of $\mathcal{P}$ and capping the INP concentration according to T_{cap} : in simulations with smaller magnitudes of $\mathcal{P}$ and higher values of T_{cap} , a larger proportion of the cloud will experience more INP limitation.

To represent ice nucleation during the case study, the INP parameterisation in the INP-limited scheme was calculated by applying the fit used in Section A.3 to an average of the upstream INP concentrations, and limiting this at T_{cap} , such that

$$N_{\text{INP}} = \begin{cases} a \exp \left[b + c(d - T_{\text{cap}}/e)^f \right], & T > T_{\text{cap}} \\ a \exp \left[b + c(d - T/e)^f \right], & T_{\text{cap}} < T < -6^\circ\text{C} \\ 0, & T > -6^\circ\text{C}, \end{cases} \quad (5.5)$$

where $a = 1000 \text{ m}^{-3}$, $b = -5.634$, $c = 1.4$, $d = -5.0$, $e = 1^\circ\text{C}$ and $f = 0.588$, and T is in units of $^\circ\text{C}$. The number of traced INPs in droplets $N_{\text{trINP}, w}$, rather than the number of insoluble aerosols in droplets, was used to limit the amount of ice nucleation, such that

$$\frac{dN_i}{dt} = \frac{\min(N_{\text{INP}}, N_{\text{trINP}, w})}{\Delta t}. \quad (5.6)$$

This parameterisation is shown in Figure 5.3 alongside the default parameterisation for comparison.

Since the INP concentration must be derived at a single temperature, two different values of T_{cap} were used: -15°C and -20°C , representing higher and lower ends of the temperatures experienced by most of the cloud system. For each of these values of T_{cap} , three model simulations were performed with different INP profiles, $\mathcal{P}$, which are shown in Figure 5.4. The simulations *Observed-15* and *Observed-20* (where the suffix represents the value of T_{cap}) use the profile of INP concentration derived from observations. We then adjust the magnitude of this profile to perform four further simulations. *Abundant-15* and *Abundant-20* use

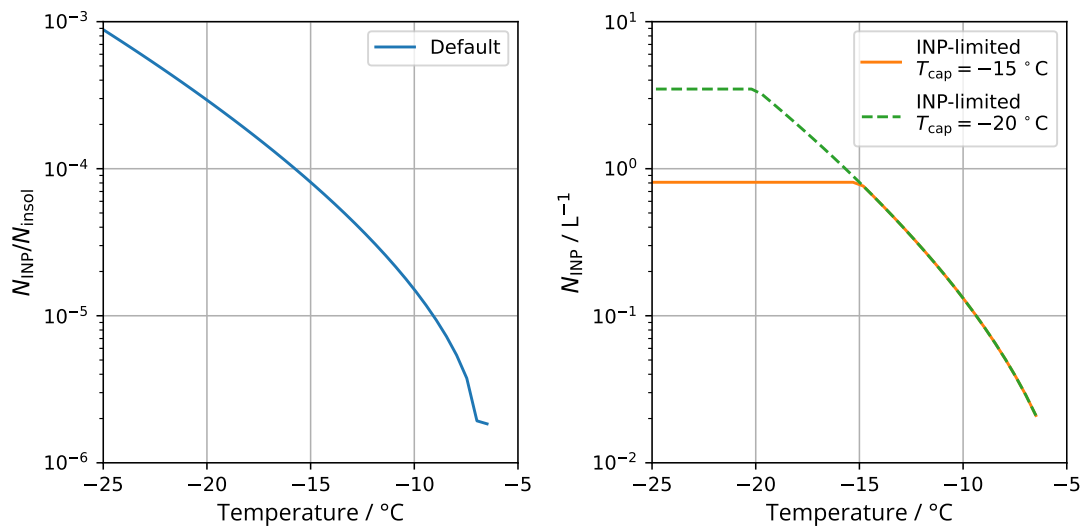


Figure 5.3: The INP parameterisations used in the default and INP-limited schemes. The INP-limited scheme is shown using the two values of T_{cap} which were used in these simulations.

an INP profile with ten times as many INPs as the *Observed* cases, representing an allegory to the *default* simulation, where INP concentrations are not strongly limited by aerosol. In contrast, *Scarce-15* and *Scarce-20* use an INP profile with ten times fewer INPs than the *Observed* cases, further suppressing the nucleation and representing an environment where INPs are largely “used up” immediately after cloud formation.

By comparing the rate of change of INP concentration in the simulations using the INP-limited scheme to the default (aerosol-limited) scheme, we can test the impact of directly modelling INPs on INP removal. If the more strongly INP-limited simulations exhibit stronger INP removal rates than the default simulation and the less strongly INP-limited simulations, it will suggest that INP removal cannot be adequately represented when active sites are untracked.

5.3 Results

Figure 5.5 shows INP concentrations at -15°C for each simulation in each of the six coloured areas used for analysis in Chapter 4, integrated below 2000 m, approximately the cloud top height at the boundary of boxes 4 and 5. In every simulation, the total column INP concentration reduces along the development of the CAO. In all of the direct simulations, the reduction in INP concentration between boxes 1 and 6 is greater than in the default simulation, but this becomes increasingly strong with increasing INP limitation, showing that the availability of INPs in the default case inhibited the removal of INPs. Values of this reduction, proportional to the concentration in box 1, are given in Table 5.2. While the reduction in the default case was 36%, the only simulation where the reduction was less than 60% was *Abundant-20* (39%), the INP-limited simulation with the weakest limitation on INP con-

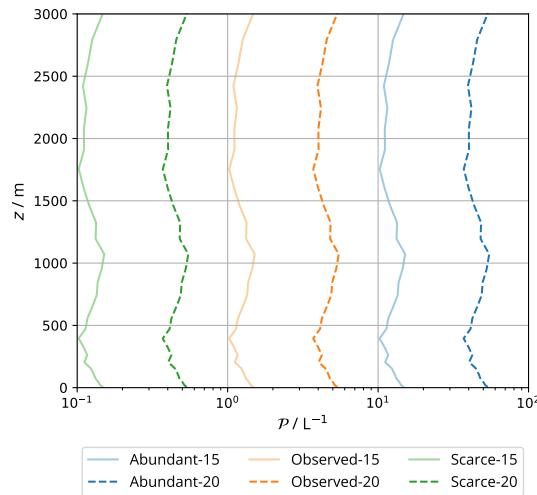


Figure 5.4: The magnitudes of the initial INP profiles, $\mathcal{P}$, supplied to the model in the *Abundant*, *Observed* and *Scarce* simulations.

centrations. As hypothesised, the strongest reductions came in the strongly INP-limited simulations, *Scarce-15* and *Scarce-20* (96 % and 95 % respectively). The more efficient INP removal in the INP-limited simulations occurred can be attributed to the reduced cloud glaciation. In these simulations, crystals were able to grow larger through the WBF process and thus precipitate more efficiently. In contrast, the increased INP concentrations in the default and INP-rich simulations resulted in more abundant, smaller crystals, which precipitated less efficiently.

While the two strongly INP-limited simulations, *Scarce-15* and *Scarce-20*, exhibited similarly strong reductions in INP concentration, in the other two pairs of simulations, the simulation using the lower value of T_{cap} had a smaller reduction in INP concentration (Table 5.2 and Figure 5.5). This difference was largest between the *Abundant-15* and *Abundant-20* simulations, which had reductions of 61 % and 36 % respectively. Such a large difference is consistent with the hypothesis that the increased availability of INP inhibits INP removal, since using Eq. 5.5, $N_{\text{INP}}(-20^\circ\text{C})$ is approximately 4 times smaller when $T_{\text{cap}} = -15^\circ\text{C}$ than when $T_{\text{cap}} = -20^\circ\text{C}$. As the *Abundant-15* and *Abundant-20* simulations had surpluses of INP at all cloud temperatures, the T_{cap} -related reduction in available INP in clouds with $T \approx -20^\circ\text{C}$ was stronger than the other simulations where INP concentrations were already limited by the magnitude of N_{trINP} .

Increasing the limitation on the INP concentration also resulted in earlier onsets in the reduction of INP concentration, as a greater proportion of the INPs were processed and removed earlier in the outbreak. Between boxes 3 and 4, the column-integrated INP concentration fell by 10 % in *Abundant-15*, 34 % in *Observed-15* and 71 % in *Scarce-15*.

The changes in the column-integrated INP concentrations are largely reflected in vertical profiles of the INP concentration, shown in Figure 5.6. As in the default case, INP concentra-

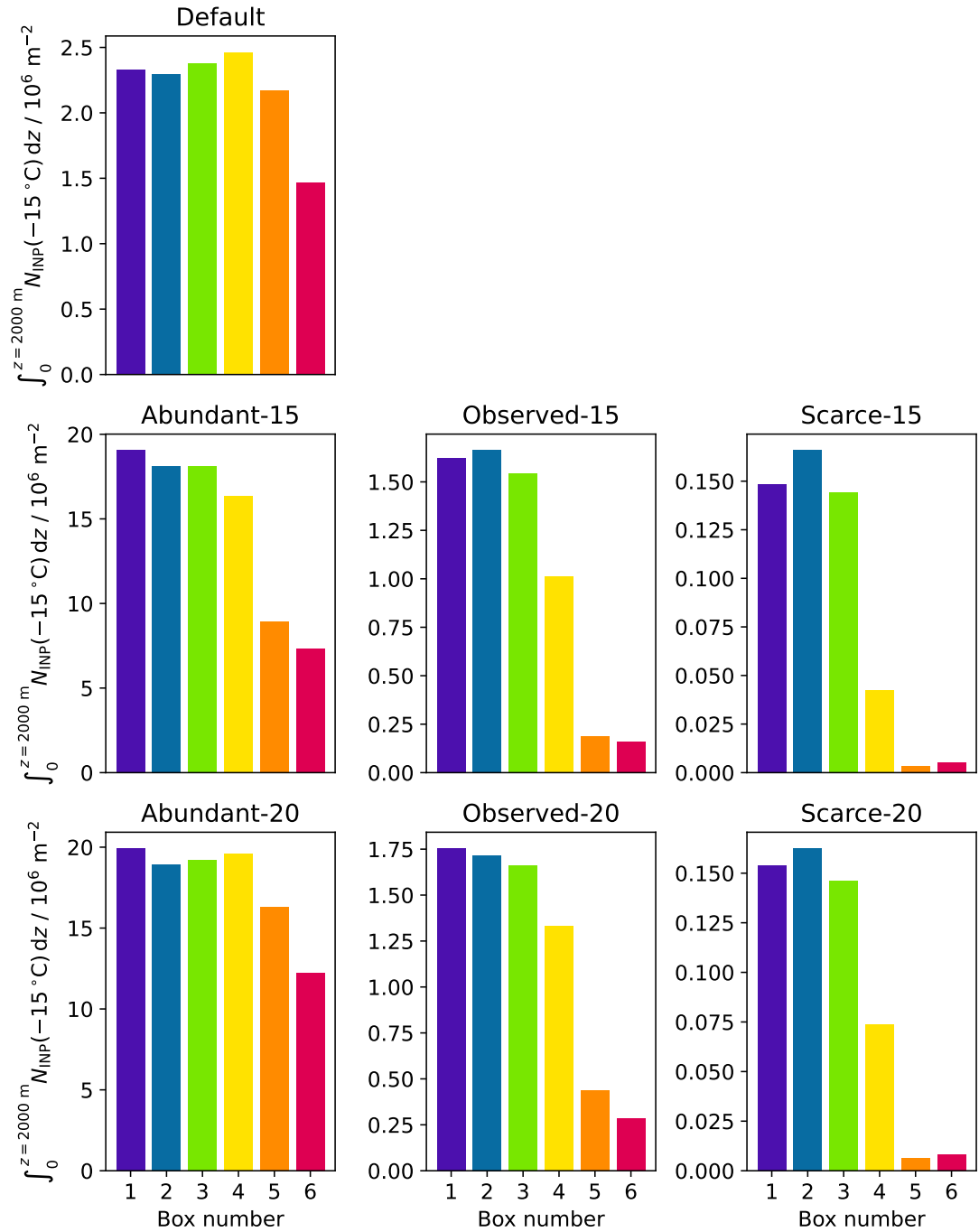


Figure 5.5: INP concentrations, column-integrated from 0 m to 2000 m in each analysis box for each simulation.

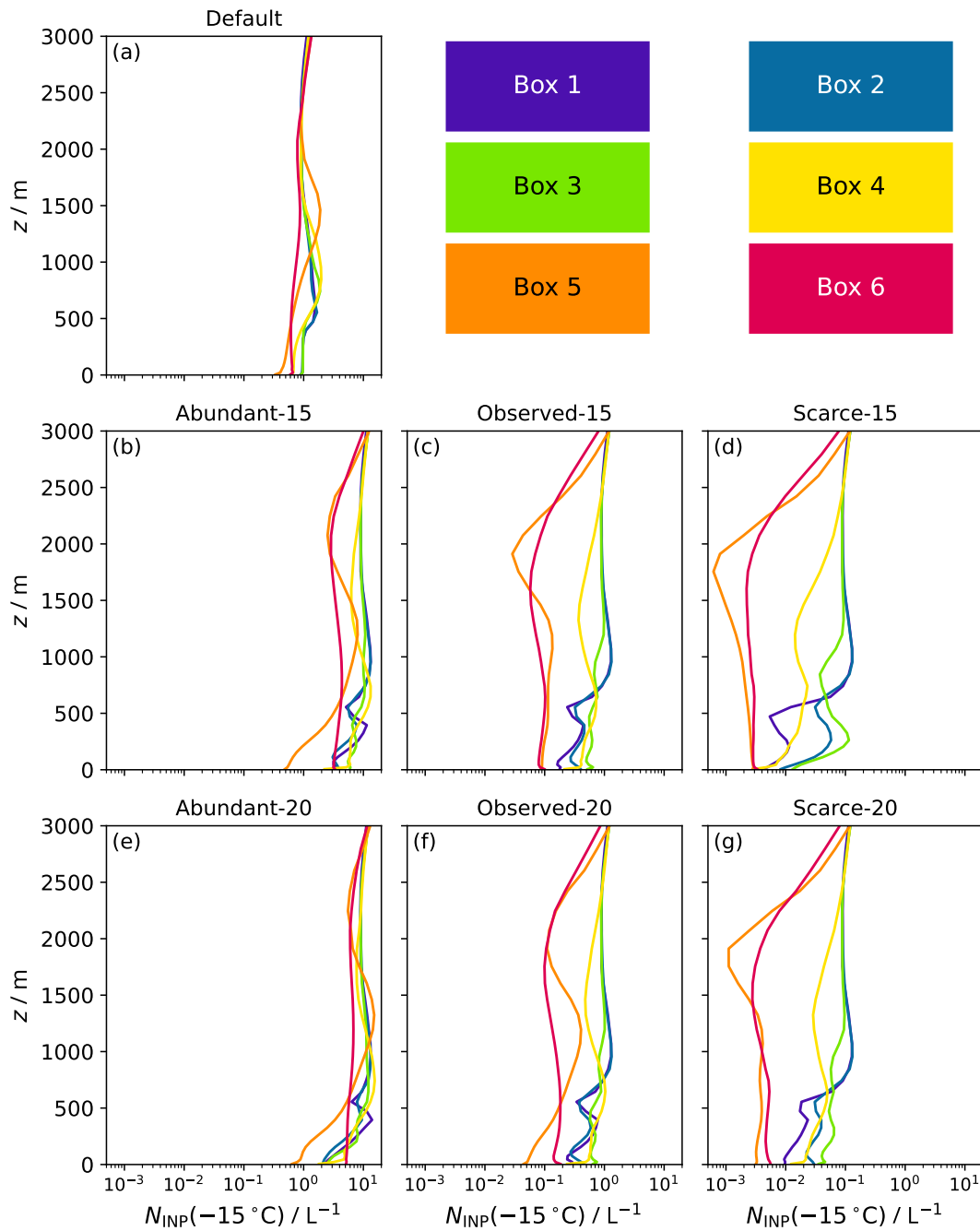
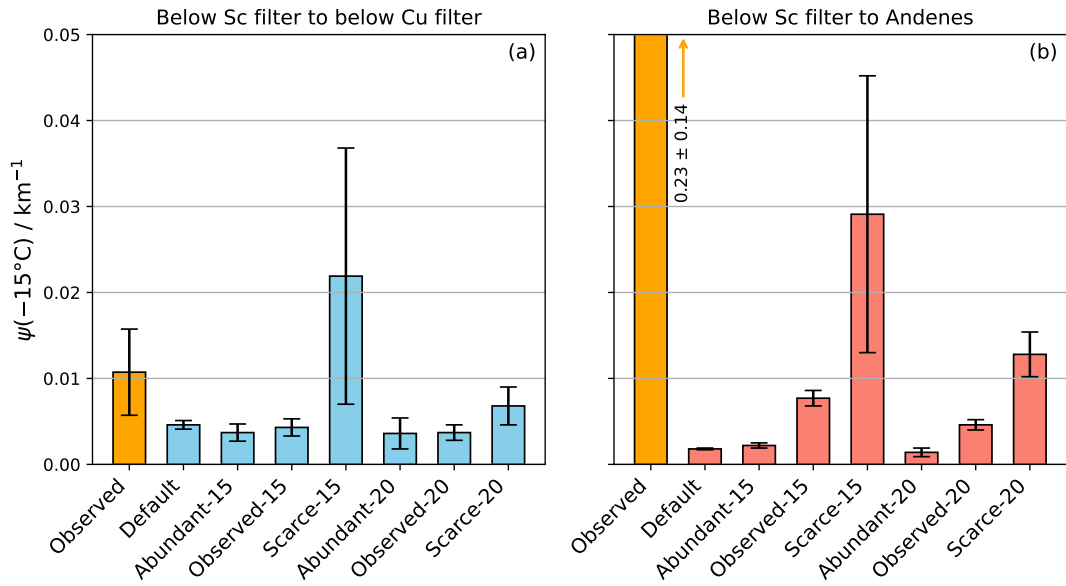


Figure 5.6: Profiles of INP concentration in each of the six coloured analysis regions shown in Figure 4.3a for each of the seven simulations. To enable easy comparison between simulations, the horizontal axes are consistent between figures. For easier comparison between profiles within a simulation, a version of this figure where the horizontal axes limits vary, Figure D.1, is provided in Section D.1.

Table 5.2: Percentage reductions in the column-integrated INP concentration at -15°C between analysis boxes 1 and 6.

Simulation	$\Delta \int_0^{2000\text{m}} N_{\text{INP}}(-15^{\circ}\text{C}) dz / \%$
Default	36
Abundant-15	61
Observed-15	90
Scarce-15	96
Abundant-20	39
Observed-20	84
Scarce-20	95

**Figure 5.7:** Values of $\psi(-15^{\circ}\text{C})$ when comparing modelled INP concentrations between locations of (a) the two below-cloud aircraft measurements and (b) the upstream below-cloud aircraft measurement and Andenes. In each case, observed values are shown in orange. The value which does not fit on the scale is shown next to the bar in question.

tions in all of the INP-limited simulations are lowest in boxes 5 and 6, though boxes 1 and 2 have lower concentrations than boxes 3 and 4 at $z < 500$ m.

In Chapter 4, we defined ψ , the reduction in INP concentration below-cloud between two locations, normalised by the distance between them, and used ψ to compare the reduction in INP concentration between the model and observations. Values of $\psi(-15^{\circ}\text{C})$ for each simulation are shown in Figure 5.7 and tabulated in Table 5.3. The two strongly INP-limited simulations, *Scarce-15* and *Scarce-20*, were able to replicate the value of $\psi(-15^{\circ}\text{C})$ observed between the two aircraft measurements to within 1σ . The ratio $\psi_{\text{obs}}(-15^{\circ}\text{C})/\psi(-15^{\circ}\text{C})$ was 0.5 for *Scarce-15* and 1.6 for *Scarce-20*, representing an improvement on the default simulation, where the ratio was 2.3. As hypothesised, $\psi(-15^{\circ}\text{C})$ increased as INP limitation increased, but with the exception of *Scarce-15*, all of the INP-limited simulations were consistent (1σ) with the default simulation. This consistency was related to the location

Table 5.3: Values of the INP concentration drop-off rate, ψ , in each of the simulations.

Simulation	Below Sc filter to below Cu filter		Below Sc filter to Andenes	
	$\psi (-15^\circ\text{C}) / \text{km}^{-1}$	$\frac{\psi_{\text{obs}}(-15^\circ\text{C})}{\psi(-15^\circ\text{C})}$	$\psi (-15^\circ\text{C}) / \text{km}^{-1}$	$\frac{\psi_{\text{obs}}(-15^\circ\text{C})}{\psi(-15^\circ\text{C})}$
<i>Observed</i>	0.011 $\pm$ 0.005	—	0.23 $\pm$ 0.14	—
Default	0.0046 $\pm$ 0.0005	2.3	0.0018 $\pm$ 0.0001	120
Abundant-15	0.0037 $\pm$ 0.0010	2.9	0.0022 $\pm$ 0.0003	100
Observed-15	0.0043 $\pm$ 0.0010	2.5	0.0077 $\pm$ 0.0009	29
Scarce-15	0.022 $\pm$ 0.015	0.5	0.029 $\pm$ 0.016	7.8
Abundant-20	0.0036 $\pm$ 0.0018	3.0	0.0014 $\pm$ 0.0005	170
Observed-20	0.0037 $\pm$ 0.0009	2.9	0.0046 $\pm$ 0.0006	50
Scarce-20	0.0068 $\pm$ 0.0022	1.6	0.0128 $\pm$ 0.0026	18

where the onset of substantial removal of INP began, which differed between each simulation. While the aircraft measurements were made between boxes 3 and 4 (green and yellow), in the model, INP removal typically began substantially in boxes 4 and 5.

Over the larger distance between the measurements and Andenes, there was a stronger increase in ψ with increasing INP limitation. Both INP-rich simulations (*Abundant-15* and *Abundant-20*) were consistent with the default simulation, while the *Observed-15* and the *Observed-20* simulations had significantly higher removal than their INP-rich counterparts and the strongly INP-limited simulations had significantly higher removal again. This adds further evidence to the hypothesis that the default simulation suppressed the removal of INP since the relationship between aerosols and INP did not evolve as active sites were untracked. However, none of these simulations showed the strength of reduction in INP concentrations suggested by the measurements at Andenes: even for *Scarce-15*, which had the strongest removal, $\psi_{\text{obs}}(-15^\circ\text{C}) / \psi(-15^\circ\text{C})$ was 8. In Section 4.4.2, we suggested that some of the discrepancy between the model and observations may have been due to the difference between the two measurement techniques, noting that discrepancies in INP concentration of an order of magnitude between techniques are not uncommon (Hiranuma et al., 2019; Lacher et al., 2024). If the discrepancy between the techniques were biased such that the measurements at Andenes were an order of magnitude too low (perhaps due to insensitivity to biological components of the aerosol), the observed value of ψ would have been consistent with modelled values in the *Observed-15*, *Scarce-15* and *Scarce-20* simulations. However, it seems likelier that the difference between the observed and modelled values of ψ are partially due to discrepancies between the techniques and partially due to the efficiency of removal in model processes independent of the ice nucleation scheme.

Figure 5.8 shows the evolution of INP concentration through the domain at $z = 1195$ m, a model level approximately central to the cloud in analysis box 4 (yellow). Additionally, Figure 5.9 shows the evolution of average in-cloud INP throughout the cold-air outbreak for each of the simulations alongside the measured INP concentrations for comparison. Although the two simulations with the strongest limitation on INP concentration (*Scarce-15*

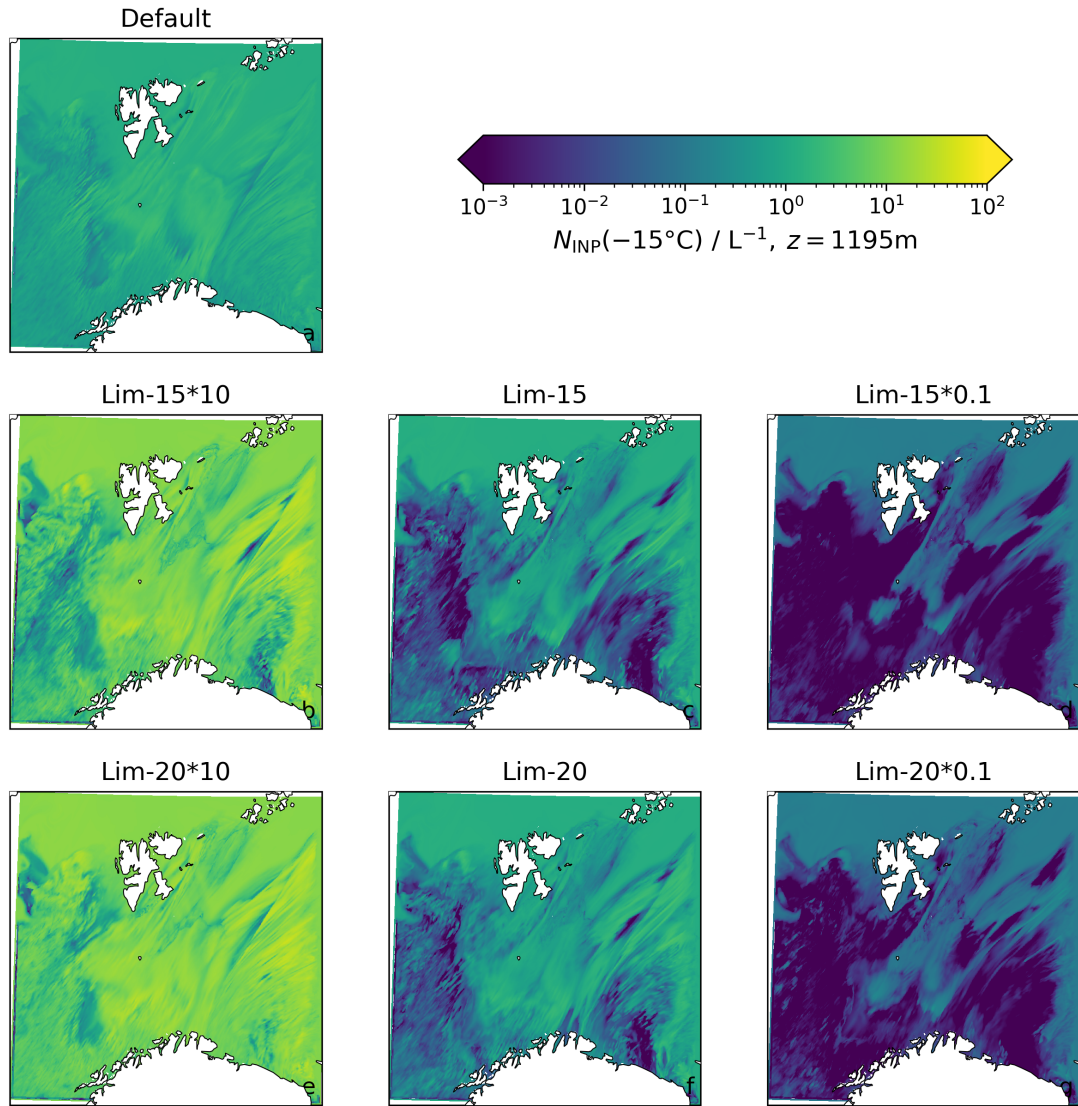


Figure 5.8: Maps of INP concentrations in each simulation at 1195 m.

and *Scarce-20*) best captured the magnitude of reduction in INP concentrations between the two aircraft observations, the magnitude of the INP concentration was too low by an order of magnitude in these simulations. No simulations exhibited INP concentrations as low as those observed at Andenes, but where simulations did well-capture the aircraft measurements of INP concentration (*Observed-15*, *Observed-20* and the default simulation), the magnitude of reduction in INP concentration could not be captured. Our inability to simultaneously replicate both the INP concentrations and the magnitude of their reduction may be due to compromises required to implement the INP-limited scheme in the UM and CASIM. As N_{trINP} , the INP tracer, was initiated according to a profile of INP concentration calculated at a specific temperature, the temperature cap on the freezing rate prevented excessive freezing in the INP-rich simulations due to lower temperatures, rather than more INPs being available. However, this cap also reduced the rate of freezing at the coldest cloud tops downstream, which reached -25°C . This would have reduced the quantity of ice pro-

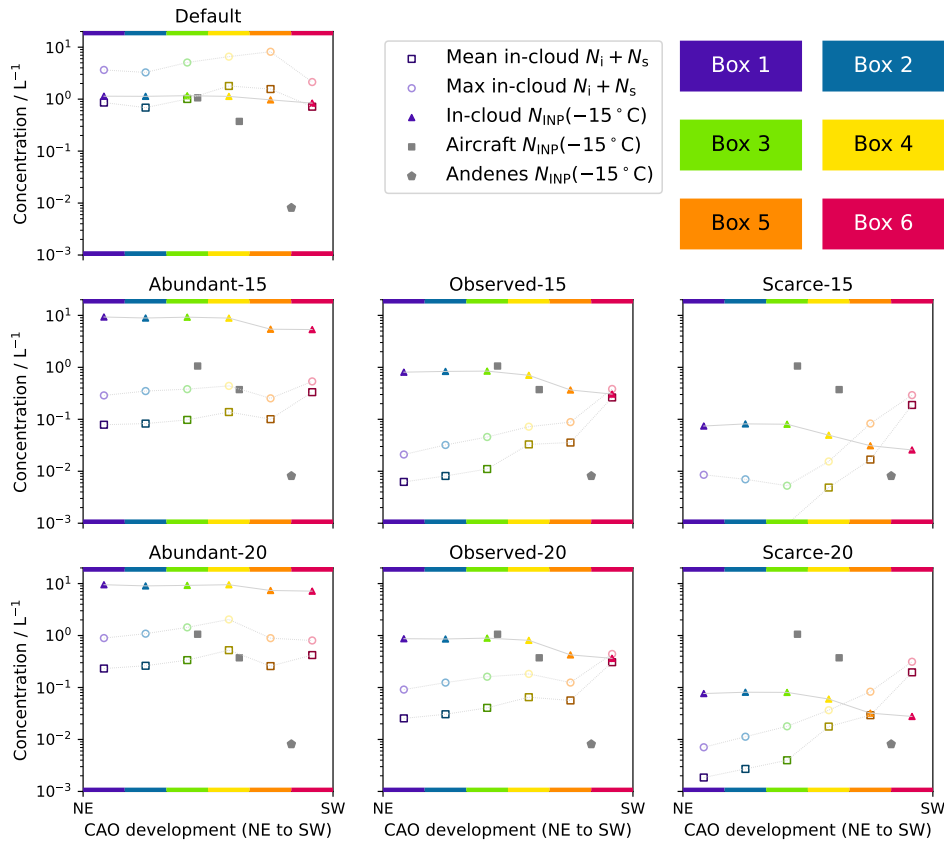


Figure 5.9: In-cloud INP concentrations compared to ice concentrations. Observational data is coloured grey for comparison, though these measurements were made below the cloud.

duction in these coldest clouds, where the extra ice produced could have otherwise acted to further accelerate the removal of INPs through enhanced precipitation.

In Chapter 2 and Chapter 4, we contrasted the observed reduction in INP concentrations through the CAO with the increase in ice concentrations observed by remote sensing instruments (Murray-Watson and Gryspeerdt, 2024). Figure 5.9 compares the evolution of in-cloud INP concentrations through the CAO to the evolution of in-cloud ice concentrations. Though INP concentrations reduced through the CAO, this was not true of in-cloud ice concentrations, which either remained approximately constant (within half an order of magnitude) or increased throughout the CAO, sometimes by nearly two orders of magnitude. Perhaps counter-intuitively, the simulations with the strongest opposite trends were those which were most strongly INP-limited. One simulation exhibiting such a large increase in the ice concentration was *Observed-15*. Figure 5.10 shows the ice and INP concentrations in this simulation alongside the development of the CAO cloud through a transect along the analysis region. As the cold-air outbreak develops along the transect, the clouds deepen

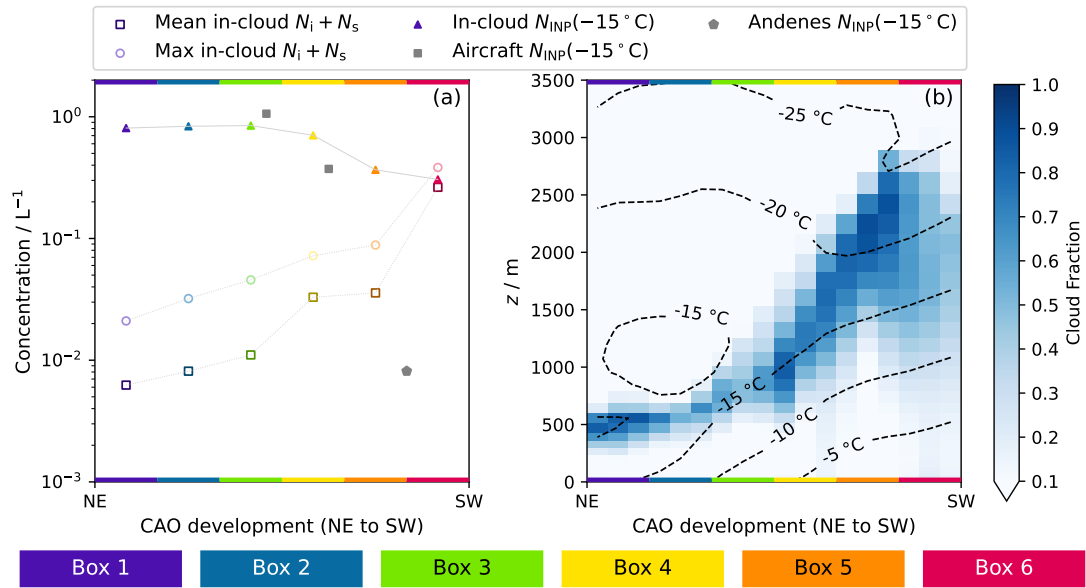


Figure 5.10: The development of the CAO in simulation *Observed-15* along the direction of the flow within the six analysis boxes. Panel (a) compares maximum and mean in-cloud ice and snow concentration ($N_i + N_s$) with the in-cloud INP concentration for each of the analysis boxes. Panel (b) shows the grid-box cloud fraction with contours of temperature overlaid.

and access lower temperatures. In these deeper clouds, ice concentrations are enhanced by freezing of lower-temperature INP, which form a larger subset of the remaining aerosol. As such, the concentration of ice increases while $N_{INP}(-15^\circ C)$ decreases.

5.4 Recommendations for future model development

In Section 5.3, we found that we were unable to simultaneously represent the correct magnitude of the INP concentration and the correct magnitude of INP removal through the outbreak. This was attributed to the role of T_{cap} , which inhibited freezing in the coldest clouds in the outbreak. This highlights a second deficiency that occurs when ice nucleation sites are assumed to be uniformly distributed among the aerosol population, which is described in Figure 5.11a, which considers a cloud at temperature T_{cl} . We might expect that INPs with activation temperatures $T_{act} > T_{cl}$ would nucleate ice, while INPs with activation temperatures $T_{act} < T_{cl}$ would remain unaffected. If the cloud parcel rose and experienced a lower temperature, we would expect significant further ice nucleation by the previously unaffected INPs with lower activation temperatures. In contrast, if the cloud parcel sank and experienced a higher temperature, we would expect little further ice nucleation, since INPs which can activate at this temperature have already nucleated, are unable to nucleate again, and may have already been removed by precipitation. In the default and INP-limited simulations, the model was unable to represent this since $f(T)$ is continuous and monotonic, which meant that N_{INP} was reduced by the same factor at all temperatures between timesteps.

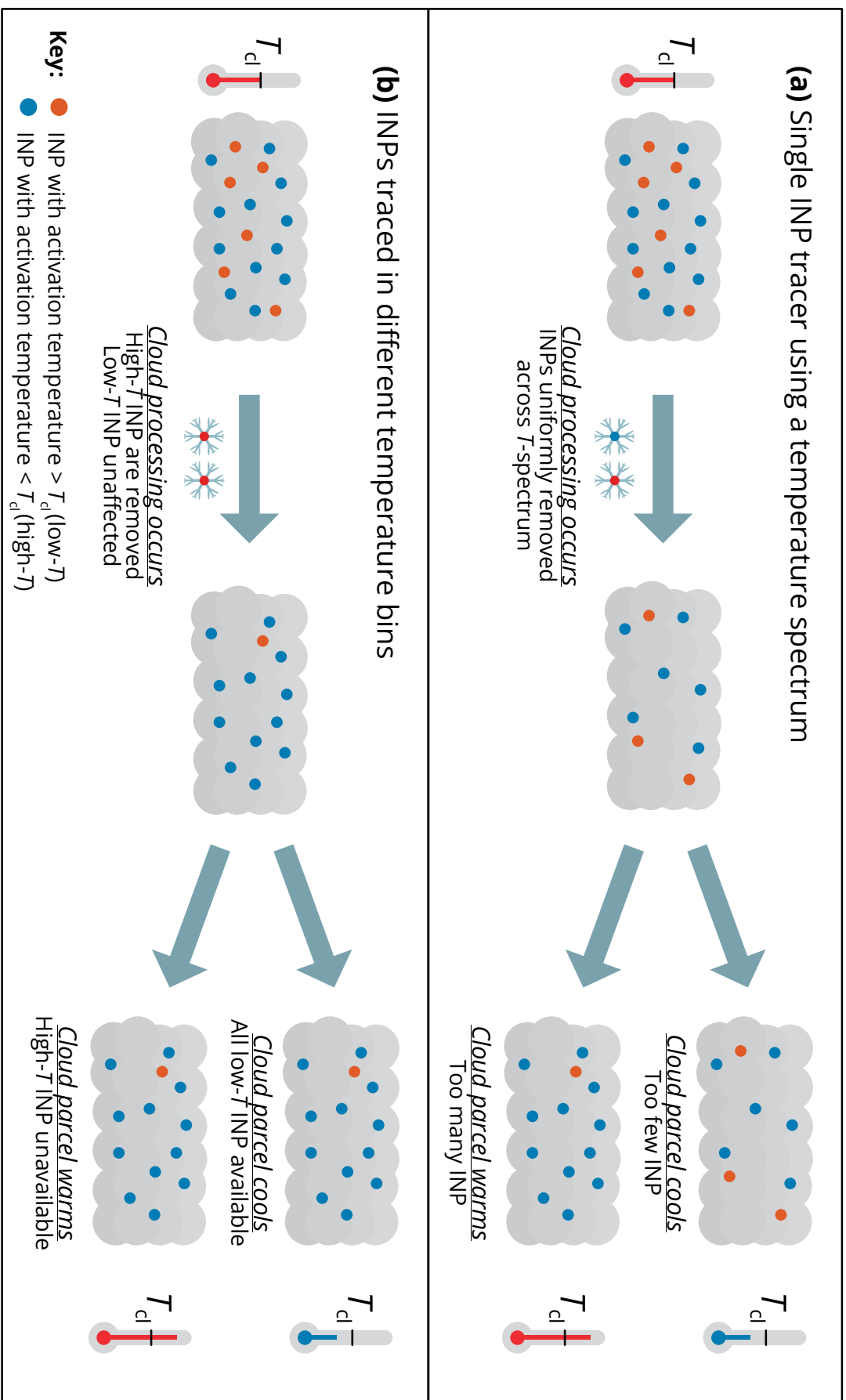


Figure 5.11: A schematic describing how the tracing INP concentrations in multiple bins determined by freezing temperature of the INP could improve the representation of INP removal.

A better, but more computationally-expensive approach is described in Figure 5.11b. Here, multiple tracers are used to trace concentrations of INP which are active in different temperature ranges. This allows INP concentrations to be limited in each temperature range while still permitting freezing to be significantly enhanced at lower temperatures. Such an approach may produce sufficient ice production to allow us to accurately model INP concentrations and removal rates simultaneously.

5.5 Conclusions

In this study, we modified the aerosol-aware implementation of ice nucleation in CASIM to perform the first regional-scale modelling of a CAO where the INP concentration is directly traced rather than assuming that the more-abundant dust aerosol acts as a very large reservoir of potential INP. We found that this assumption artificially inflates the INP concentration, and therefore underestimates the reduction in INP concentration through the cloud system as INPs are removed in precipitation.

In simulations where the clouds were strongly INP-limited, we were able to replicate the observed magnitude of the reduction in INP concentrations, but we were unable to replicate both the INP concentrations and the magnitude of their reduction at the same time. Further model development which bins INPs according to the temperature they freeze at may offer an opportunity to achieve this.

Though this modelling confirms that INP concentrations reduce through a cold-air outbreak, both modelling and assessing the magnitude of such a change is difficult to perform as it relies on measurements throughout the cloud system, which do not exist. While the aircraft measurements that were available suggested a reduction in INP concentration, their spatial proximity meant that our attempt to replicate that reduction in the model was also strongly sensitive to the location where reductions in INP concentration began. The location of the onset of INP removal through precipitation may have been sensitive to a large number of cloud-controlling factors. The aircraft and Andenes measurements were separated by a much greater distance, reducing this sensitivity. However, there were differences between the filter technique used to sample INPs on the aircraft and the impinger technique used to sample INPs at Andenes. This meant it was difficult to quantify how much of the discrepancy in the reduction of INP concentration between model and observations could be attributed to model processes insufficiently removing INPs, and how much could be attributed to differences in the experimental techniques used to make the observations. We recommend that future planners of field campaigns measuring CAOs attempt to measure INPs along the full extent of the cloud system: this was impossible during ACAO due to airspace restrictions and fuel capacity.

Chapter 6

Conclusions

This thesis has used in-situ observations and modelling to explore the role of ice-nucleating particles in cold-air outbreaks, mixed-phase cloud systems which are critical to the cloud-phase feedback.

In Chapter 2, we described measurements of INPs made in Arctic CAOs to establish their concentrations and origins. A case study from this campaign suggested that INPs may be efficiently removed by precipitation as the CAO developed. To investigate this, in Chapter 4, we used the UM with a new two-moment microphysics scheme that used interactive aerosol to investigate the extent to which INP concentrations reduce as a CAO develops. In doing so, we identified a potential model deficiency in typical aerosol-aware ice nucleation schemes, whereby INPs are represented as a function of a larger aerosol population which does not evolve after ice nucleation, meaning that active sites are untracked. We investigated the impact of this model deficiency on INP removal in Chapter 5. A summary of the key results is provided below, followed by concluding remarks on the implications of these findings and suggestions for future research.

6.1 Summary of results

High INP concentrations associated with Arctic haze in springtime CAOs

- In March 2022, we performed the first airborne INP measurements targeted at the CAOs over the Norwegian and Barents seas.
- The INP concentrations we measured were among the highest ever observed in the Arctic, ranging from 0.18 L^{-1} to 2.1 L^{-1} when the CAO had a strong northerly component.
- The dominant INPs were most likely to be long-lived biologically enriched dust aerosol from long-range transport mechanisms most commonly associated with Arctic haze.

Evolution of INP concentration in an Arctic CAO

- We performed the first interactive, aerosol-aware simulation of INPs in a CAO to model a case study from the ACAO campaign which showed evidence that CAOs efficiently removed INPs.
- The simulation showed that, when considered at a reference temperature, INP concentrations reduced downstream in a CAO, though the rate of reduction in INP concentration in the model was 2.5 times smaller than observed.
- Although aerosol which contained INPs was removed by the CAO, the removal was small enough that at cloud temperature, a steady INP concentration was maintained through the development of the CAO as the deeper clouds downstream accessed lower temperatures.

Tracking active sites matters when modelling the INP removal rate in a CAO

- We performed the first regional-scale modelling of a CAO where the INP concentration was directly traced rather than assuming that more-abundant aerosol acts as a reservoir of INP.
- By directly tracing INPs, we were able to replicate the observed magnitude of reduction in INP concentrations through a CAO. However, we were unable to replicate the observed INP concentrations and the observed reduction in INP concentrations at the same time.
- We showed that a typical aerosol-aware INP scheme which failed to track active sites and assumed a constant relationship between the amount of aerosol and the INP concentration failed to represent the removal of INPs by precipitation and artificially inflated the INP concentration during the development of a CAO.

6.2 Implications and recommendations for future work

Synthesising the work in Chapter 2, Chapter 4 and Chapter 5, this thesis represents the first work that has traced the life-cycle of INPs through a high-latitude mixed-phase cloud system. As such, the findings of this thesis have several implications for the study of INPs in these clouds.

First, these results reinforce the importance of both making in-situ observations of INPs and of targeting these observations at specific cloud systems and locations. The high INP concentrations observed during the ACAO campaign initially came as a surprise given the much lower concentrations measured in previous Arctic field campaigns. Had we attempted to model CAO clouds assuming that these existing observations were climatologically representative of CAOs, we would have greatly struggled to reproduce observations of the

clouds when using the model. Many of these previous measurements were ground- or ship-based, so this work also exhibits the importance of measurements of the vertical profile of INPs, which only airborne measurements can currently provide. Additionally, these findings consistently contradicted inferences made by remote sensing methods which observe bulk cloud properties. Barring a paradigm shift in remote sensing technology, it is unlikely that remote sensing will be able to characterise INP concentrations as well as in-situ measurements.

However, while this work shows that in-situ measurements of INPs are important, it also suggests that in-situ measurements may be of limited use if the sampling strategy is not designed to represent the cloud system as a whole — in fact, measuring INPs at a single location is unlikely to tell the story of a large-scale cloud system. For instance, if the airborne measurements presented in Chapter 2 had not existed and only the ground-based measurements at Andenes were available, we would have underestimated the INP concentration further north by multiple orders of magnitude (even accounting for differences in the experimental techniques). Such a large underestimate would have made it significantly more difficult to model the clouds in the north of the CAO in our case study. Furthermore, given that INP concentrations at Andenes were so much lower than the aircraft INP concentrations, we might even have incorrectly assumed that the dominant INPs in the CAOs were sea-spray aerosols rather than fertile dusts in the absence of composition data. Alternatively, if we had only measured at the northern extent of the outbreaks, perhaps at a ground-based station on Svalbard, we would not have observed the removal of INPs and would have had no evidence to doubt that the model assumption that INPs are uniformly distributed among the aerosol is sufficient to model CAOs.

Given these implications, we recommend that future airborne field studies attempting to characterise a mixed-phase cloud system attempt to make measurements of INPs over as much of the cloud system as possible. An ideal way to do this with a research aircraft would be to combine an offline technique such as the drop-on filter technique used in Chapter 2 with an online INP measurement technique such as the Portable Ice Nucleation Experiment, which could continuously measure INPs across a small temperature range with a high time resolution, including in-cloud (Möhler et al., 2021). The high temporal resolution of the online technique would allow for a greater quantity of measurements of INP concentration through the cloud system, and hopefully allow for a statistically-significant assessment of changes in INP concentration. This would complement the offline filter measurements, which are able to measure INPs across a much larger temperature spectrum and have increased sensitivity to high-temperature INPs, but permit lower temporal resolution and can only be used out-of-cloud. Chapter 4 showed the importance of making measurements of INPs across a wide range of temperatures, given that the decrease in temperature in the tallest clouds resulted in increases in INP concentration that counteracted the removal of INPs.

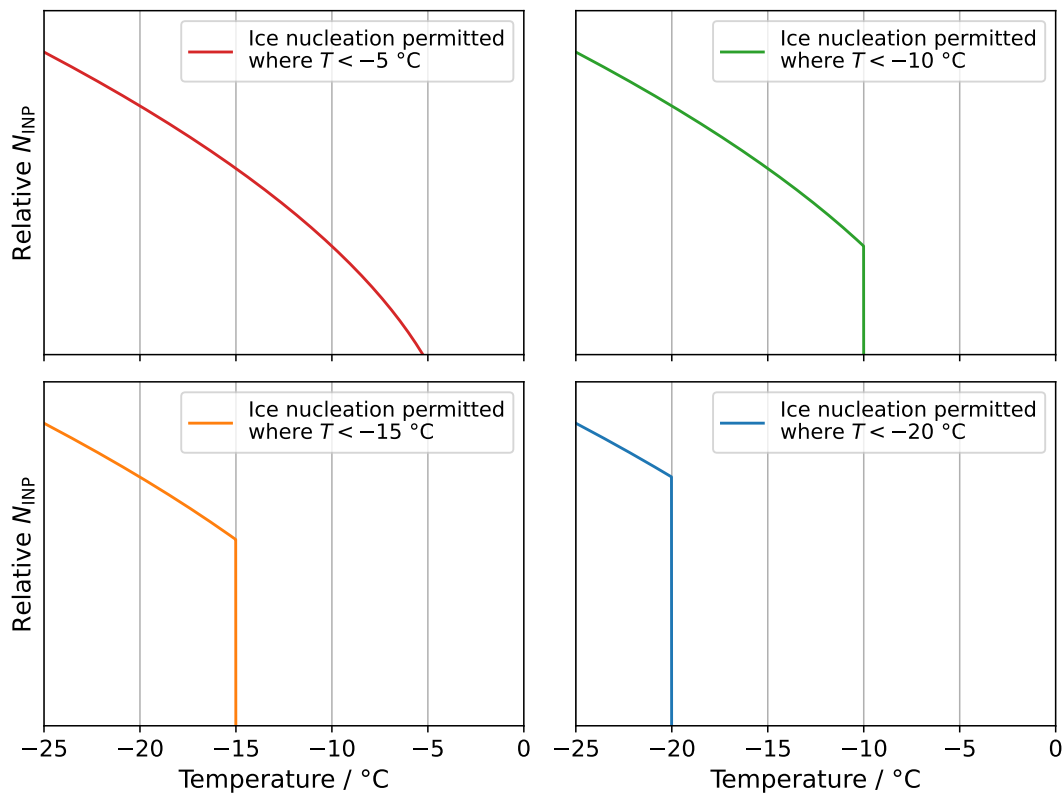


Figure 6.1: A suggested sensitivity study that could test whether tracing INPs in different temperature bins could offer improved modelling of INP removal. Each panel represents the same INP parameterisation, with freezing prohibited at progressively lower temperatures.

In Section 5.4, we highlighted a potential model deficiency whereby INPs are currently removed uniformly across the temperature spectrum, and suggested that INPs could instead be traced in different temperature bins to resolve this deficiency. While this would be highly computationally-expensive, Figure 6.1 describes a sensitivity experiment beyond the scope of this work which could test whether a bin model of INPs would be likely to improve the representation of INP removal. In this test, an INP-limited parameterisation would be used as in Chapter 5. However, freezing would be prohibited at temperatures above a varying threshold temperature, simulating the removal of progressively higher-temperature INPs. If changing the threshold temperature significantly affected the INP removal rate despite the fact INP concentration decreases with increasing temperature, it would suggest that higher-temperature INPs dominate the removed INPs and modelling INPs in temperature bins could provide an improvement on the existing model.

Finally, the larger-scale motivation for the work in this thesis was the grand challenge of improving future climate projections by reducing uncertainty on the cloud-phase feedback, which will be moderated by INPs. To do so, this work aimed to better understand the role of INPs in present-day clouds like CAOs which are susceptible to this feedback, contributing to a better representation of the clouds in models which can be used to calculate the strength

of the feedback. The findings in this thesis show that across a single cloud system, the efficient removal of INPs may mean that for some CAOs, particularly where clouds are relatively shallow and removal is especially strong, there will be significantly different cloud-phase feedback strengths in the stratocumulus and cumulus regimes owing to the reduction in INP concentration. Therefore, we suggest that when simulating the cloud-phase feedback in CAOs, INP parameterisations should ideally be interactive in order to adequately represent the removal of INPs. If this is prohibitively computationally expensive, aerosol-aware parameterisations of INP might not provide much advantage over aerosol-unaware parameterisations as they fail to capture INP removal.

Appendix A

Appendices to Chapter 2

A.1 CAO Index values

Figure A.1 shows the CAO index for each the eight flying days using average hourly ERA5 data during the flight durations. To calculate this, Eq. 1.17 was used with $p = 800$ hPa, such that

$$M_{800} = \theta_{\text{skt}} - \theta_{800}. \quad (\text{A.1})$$

Like Fletcher et al. (2016a), we chose $p = 800$ hPa since it is biased towards the detection of high-latitude CAOs. CAO indices were stronger later in the campaign, with the strongest CAOs probed during flights c275 to c279, and those with southernmost extent being probed during flights c280 to c282. Although c273 and c274 were probed in regions of more moderate CAO index, the cloud structures observed clearly resembled those of CAOs. This was also true of c271, where the CAO index was always negative. Noting the weakness of the CAO index discussed in Section 1.5.3, we conclude that these measurements were all made in CAO conditions, but that those from c275 onwards were strongest.

A.2 INP spectra and aerosol size distributions

Figure A.2 and Figure A.3 show individually labelled INP spectra and aerosol size distributions respectively, akin to Figure 2.3 and Figure 2.5 where they are grouped by location relative to the cloud.

A.3 INP spectra curve fitting

Our measurements show that the entire INP spectrum ($N_{\text{INP}}(T)$) varied over the campaign (Figure 2.3). To represent the INP concentration across the full temperature spectrum in future modelling work, particularly when modelling individual case studies, we fitted all

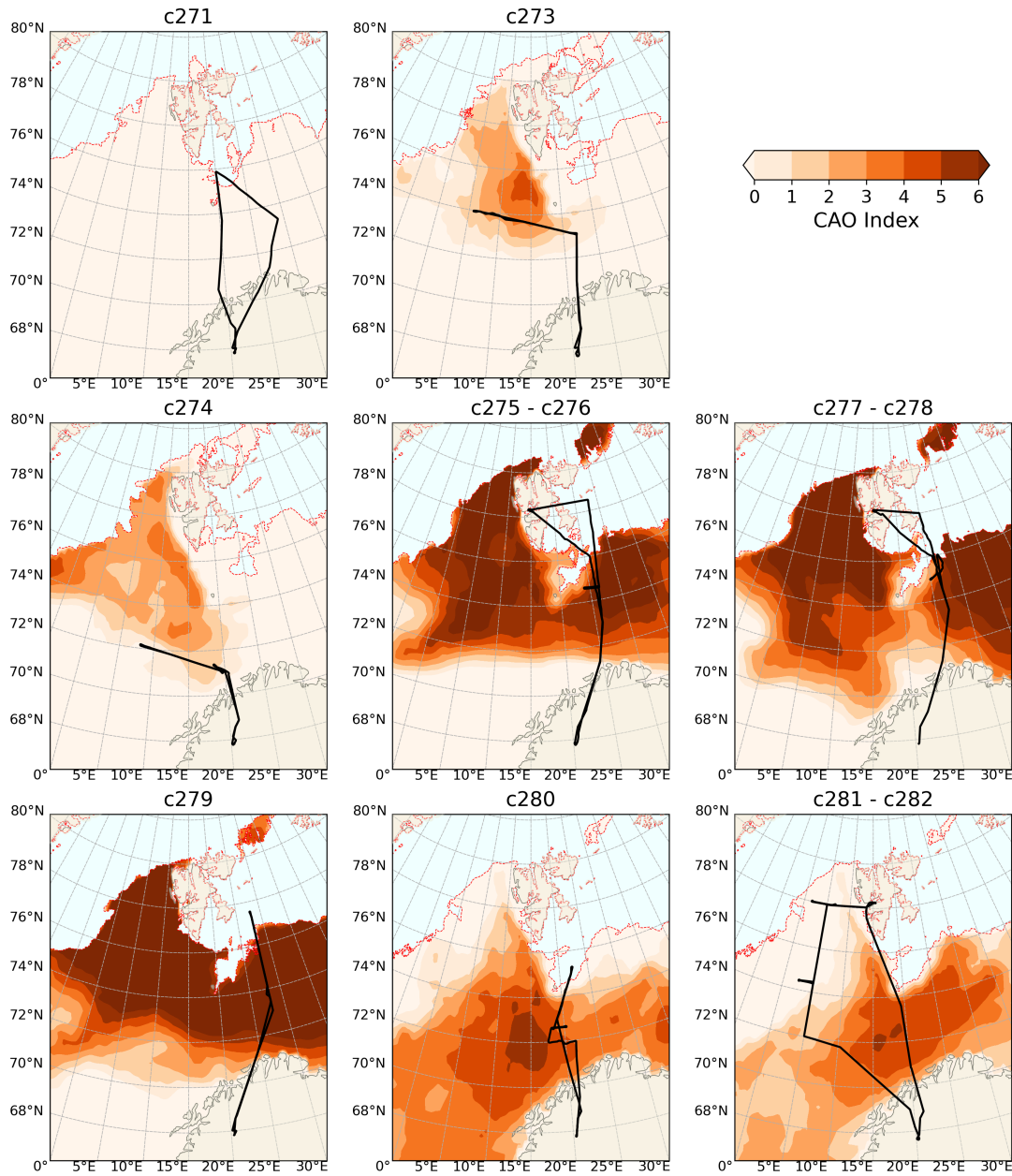


Figure A.1: CAO index of each flight, calculated using ERA5 reanalysis data for each of the eight CAO flying days (C3S, 2018). Flight tracks are overlaid in black. The CAO index is not shown over sea ice, where it is invalid.

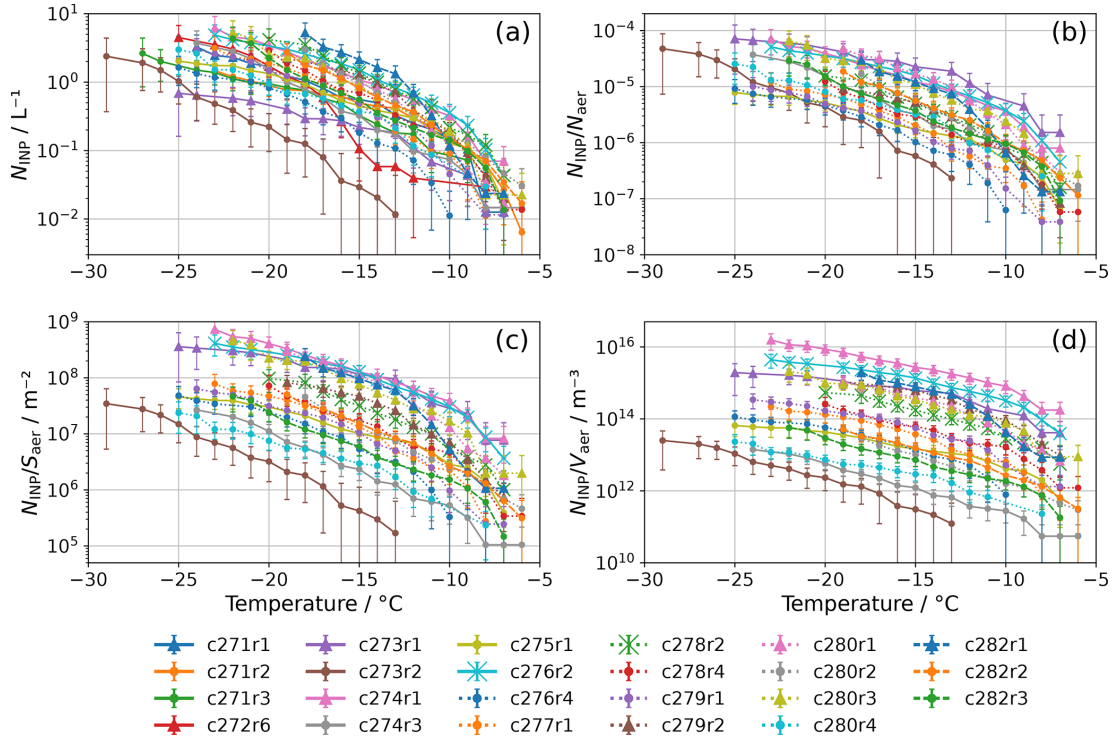


Figure A.2: Panel (a) shows INP concentrations for each sample taken during the campaign. Panels (b), (c) and (d) show these concentrations normalised by the total number (N_{aer}), surface area (S_{aer}) and volume (V_{aer}) of aerosols with diameters greater than $0.1 \mu\text{m}$.

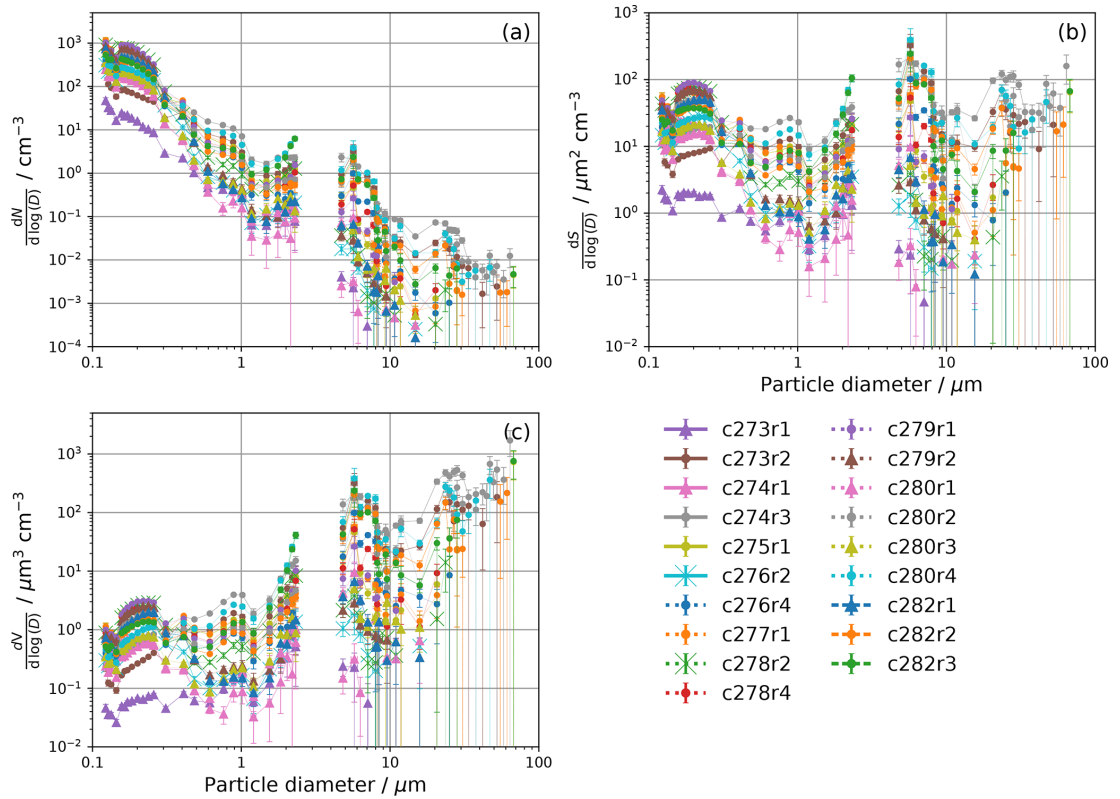


Figure A.3: Size distributions of (a) particle number, (b) particle surface area and (c) particle volume measured with the PCASP and CDP probes during each filter sample. Probe data were unavailable during flights c271 and c272.

spectra to a four-parameter function. This function took the form

$$N_{\text{INP}} = a_{\text{INP}} \exp \left[b (c - T)^d \right]. \quad (\text{A.2})$$

The parameter c represents the greatest temperature at which a freezing event was observed, b and d are shape parameters that determine the curvature of the function, and a_{INP} is a scale parameter related to INP concentrations (if the shapes were constant). For INP spectra with the same shape, greater values of a_{INP} indicate greater INP concentrations across the entire temperature range. The same functional form was also applied to active site density spectra, which were fitted to the equation

$$N_{\text{INP}}/S_{\text{aer}} = a_{\text{S}} \exp \left[b (c - T)^d \right]. \quad (\text{A.3})$$

Fitting parameters for all spectra are given in Table A.1, while examples of this fitting are shown in Figure A.4, where functions for three individual samples, c273r2, c277r1 and c276r2, are illustrated. These represent filter measurements with low, medium and high INP concentrations and active site densities and demonstrate the ability of the function to represent all INP concentrations during the campaign. Eq. A.2 and Eq. A.3 were also used to create parameterisations for the median INP concentration and active site density across the campaign for springtime CAOs in the region (excluding c272r6, which was not taken in cold-air outbreak conditions). These are also shown in Figure A.4.

Table A.1: Values of parameters that describe INP concentration (N_{INP}) and INP concentration normalised by surface area ($N_{\text{INP}}/S_{\text{aer}}$) using Eq. A.2 and Eq. A.3 respectively. There is no parameterisation of $N_{\text{INP}}/S_{\text{aer}}$ for flights c271 and c272 due to a lack of aerosol size data. Similar parameterisations for INP concentration normalised by N_{aer} and V_{aer} , not presented here, can be found with other metadata at <https://doi.org/10.5281/zenodo.11221599>.

Sample	N_{INP}				$N_{\text{INP}}/S_{\text{aer}}$			
	a_{INP} (10^{-2} L^{-1})	b	c ($^{\circ}\text{C}$)	d	a_{S} (10^6 m^{-2})	b	c ($^{\circ}\text{C}$)	d
c271r1	2.524	0.843	−7.00	0.619	–	–	–	–
c271r2	1.964	0.975	−6.00	0.531	–	–	–	–
c271r3	1.760	1.050	−7.67	0.525	–	–	–	–
c272r6	0.492	0.607	−6.00	0.849	–	–	–	–
c273r1	1.087	1.031	−6.96	0.500	5.692	1.031	−6.96	0.500
c273r2	1.156	0.616	−13.00	0.793	0.197	0.533	−13.00	0.836
c274r1	3.368	1.050	−6.64	0.570	3.993	1.050	−6.64	0.570
c274r3	0.415	0.606	−4.00	0.818	0.193	0.311	−6.00	0.960
c275r1	2.484	0.972	−7.00	0.539	0.596	0.933	−7.00	0.548
c276r2	5.088	1.029	−7.00	0.553	4.346	1.030	−7.00	0.552
c276r4	1.557	1.032	−10.00	0.570	0.462	1.031	−10.00	0.570
c277r1	1.690	0.978	−8.00	0.631	0.587	0.757	−8.00	0.652
c278r2	4.343	0.885	−7.00	0.654	1.011	0.960	−7.00	0.652
c278r4	1.095	1.050	−5.97	0.619	0.273	1.050	−5.97	0.619
c279r1	1.108	1.050	−7.00	0.605	0.269	0.960	−7.00	0.629
c279r2	2.364	1.003	−7.00	0.640	0.832	1.049	−7.00	0.626
c280r1	1.556	1.050	−7.00	0.653	1.665	1.050	−7.00	0.653
c280r2	3.160	0.474	−6.00	0.903	0.477	0.474	−6.00	0.903
c280r3	1.320	1.050	−5.87	0.624	1.163	1.050	−5.87	0.624
c280r4	1.291	1.050	−7.44	0.563	0.163	0.834	−7.81	0.617
c282r1	2.188	1.050	−7.00	0.701	0.991	1.050	−7.00	0.701
c282r2	1.223	1.050	−5.87	0.630	0.226	1.050	−5.87	0.630
c282r3	3.707	0.426	−7.00	0.880	0.368	0.472	−7.00	0.847

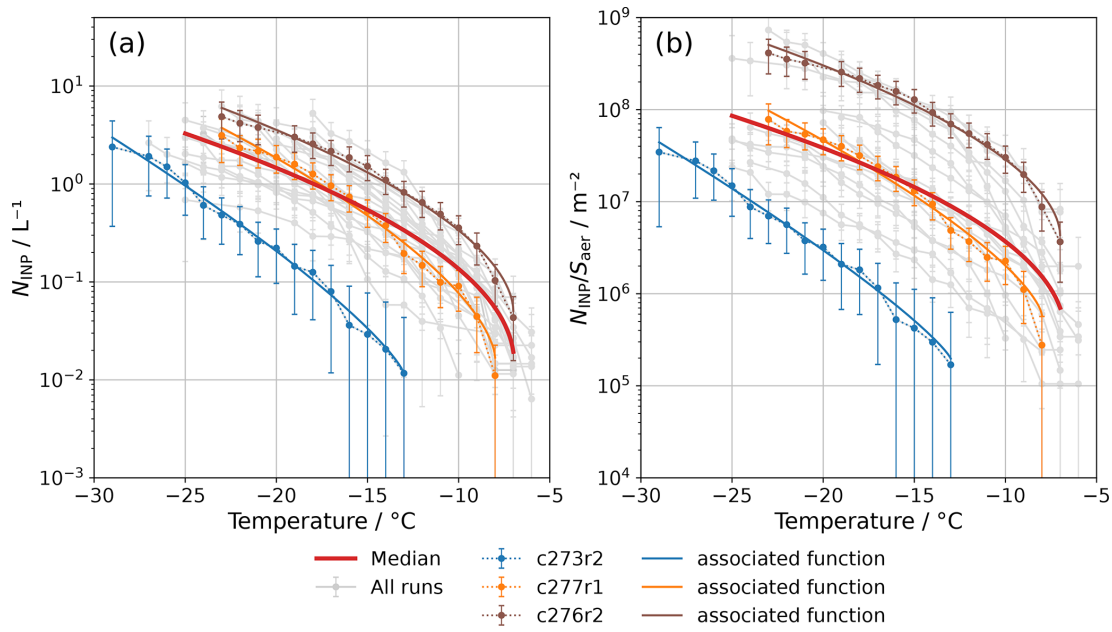


Figure A.4: All measured temperature spectra for (a) INP concentration and (b) INP concentration normalised by aerosol surface area were fitted to Eq. A.2 and Eq. A.3 respectively. In both panels, examples of fits to randomly chosen spectra with relatively high, medium and low INP spectra are demonstrated. In panel (a), the red line shows the function fitted to the median INP concentration observed during the campaign, where $a_{\text{INP}} = 1.480 \times 10^{-2} \text{ L}^{-1}$, $b = 1.271 \text{ K}^{-d}$, $c = 266.2 \text{ K}$ and $d = 0.5$. In panel (b), the red line shows the function fitted to the median active site density observed during the campaign, where $a_{\text{S}} = 3.712 \times 10^5 \text{ m}^{-2}$, $b = 1.273 \text{ K}^{-d}$, $c = 266.4 \text{ K}$ and $d = 0.5$.

Appendix B

Appendices to Chapter 3

B.1 Glossary of terminology relating to INP parameterisations

Table B.1: Glossary of terminology relating to INP parameterisations.

Term	Definition
Aerosol-aware	An aerosol-related quantity, such as number concentration, is used to calculate the INP concentration. Aerosol-aware INP parameterisations may be passive or interactive .
Aerosol-unaware	No quantities related to aerosol are used to calculate the INP concentration. By definition, aerosol-unaware INP parameterisations are passive .
Interactive	INP concentration is prognosed based on a property of the aerosol size distribution and environmental variables such as temperature. The amount of aerosol in liquid, ice and air is tracked and aerosol is moved between tracers according to the nature of the freezing, e.g. for immersion freezing, INPs are moved from the in-droplet to in-ice tracer. The opposite of passive .
Passive	INP concentration is diagnosed based on environmental variables such as temperature and possibly a property of the aerosol size distribution. If aerosol is used for calculating INP concentration in a passive simulation, it is not processed and aerosol does not move to the in-ice tracer. The opposite of interactive .

Appendix C

Appendices to Chapter 4

C.1 Comparing aircraft and model meteorology

Figure C.1 shows comparisons between the horizontal wind and relative humidity measurements made on the aircraft. Modelled wind speeds are typically within 1 m s^{-1} to 2 m s^{-1} of the measured value, with the westward component of the wind generally slightly too strong. Humidities are well represented in Profiles 1 and 2, though are overestimated in Profile 3, where the bias in ice and water contents is strongest towards ice.

C.2 Comparing aircraft data to the nearest model grid box

Figure C.2 shows a comparison between the modelled and measured temperature, cloud droplet and snow concentrations and liquid and ice water contents. Temperature measurements were made with the Rosemount platinum loom resistance thermometer (Price, 2022). Other measurements were made as described in Section 4.4.1, except values in cells with cloud fractions lower than 0.1 are not discarded.

C.3 Maps of INP concentrations at grid-box temperature

Figure C.3 shows the INP concentration at the grid-box temperature at the three cloud-relevant heights used in Figure 4.8.

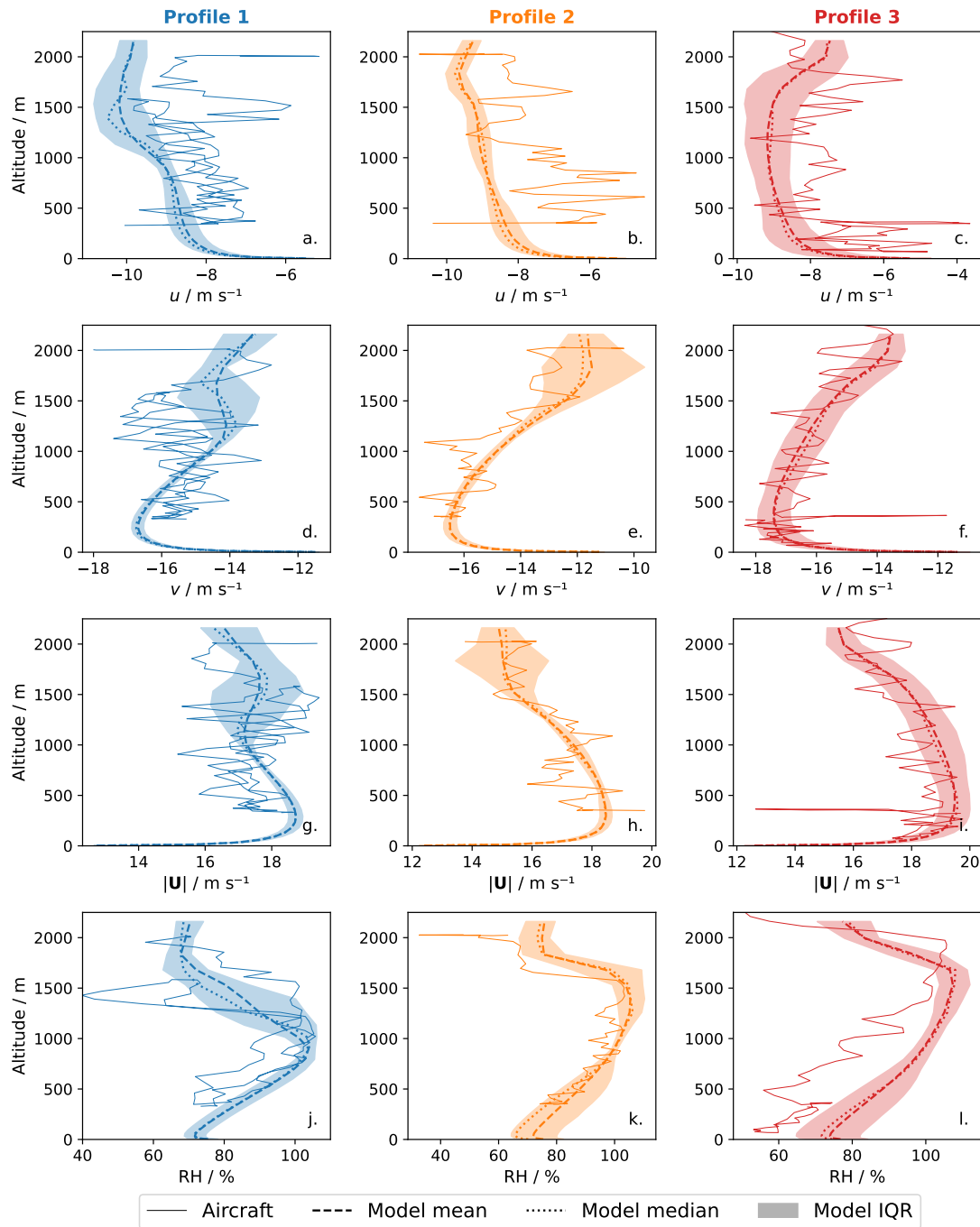


Figure C.1: Profiles of meteorological measurements made by the FAAM aircraft compared to the model. As in Figure 4.5, each column represents a different cloud profile made by the aircraft. The top row shows measurements of eastward bulk wind velocity u , while the second row shows measurements of southward bulk wind velocity, v . The third row shows the magnitude of the resultant horizontal wind vector, $|U|$. All were derived from the POS AV 410 GPS-aided Inertial Navigation unit and corrected to account for turbulence. The bottom row shows measurements of relative humidity derived from the Water Vapour Sensing System II and Rosemount probes. Aircraft measurements are averaged over 10 s, which is approximately the time taken for the aircraft to traverse a grid box. Shaded areas denote the interquartile range (IQR) of model data.

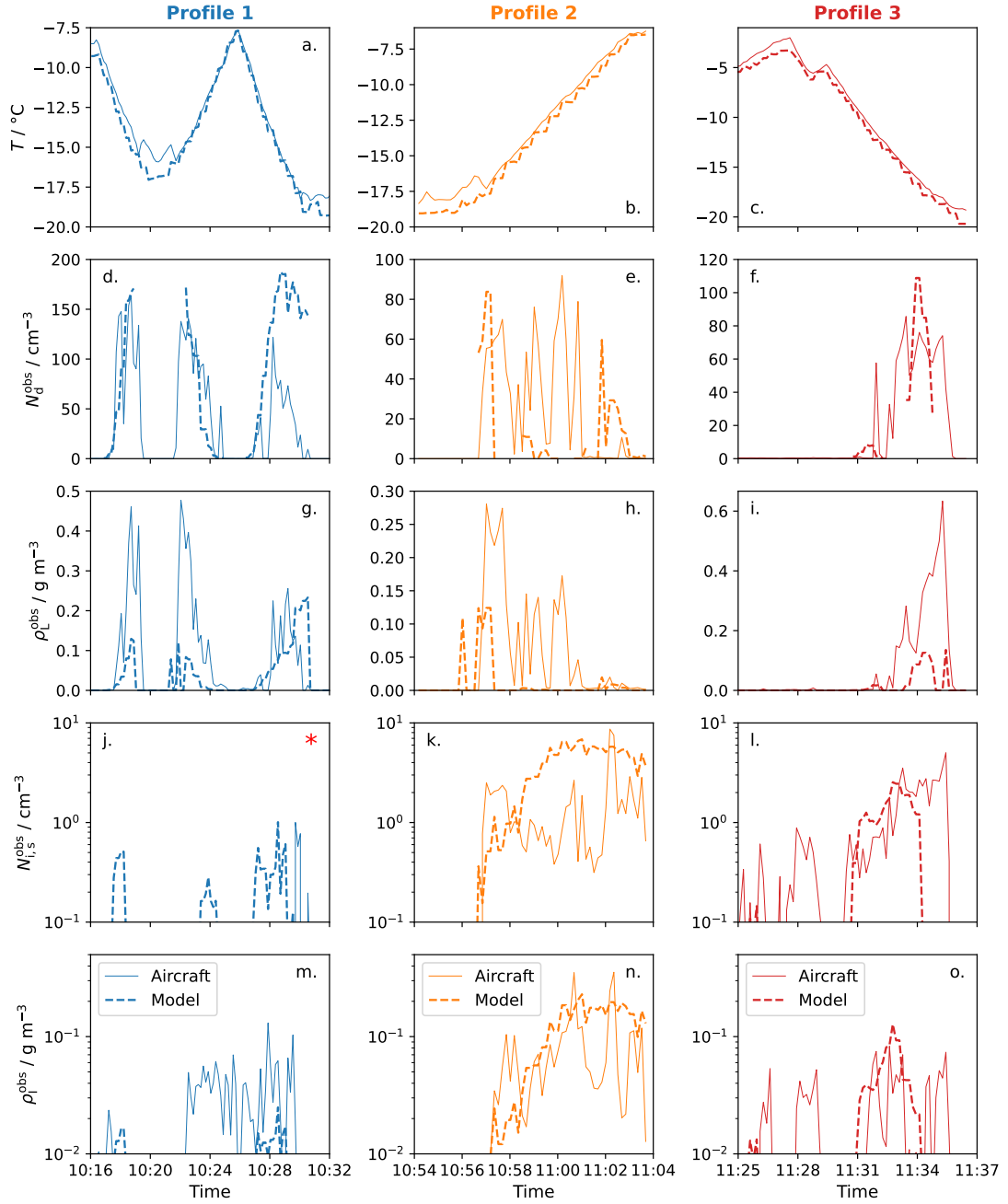


Figure C.2: A comparison of aircraft data to values in the nearest model grid box. As in Figure 4.5, each column represents a different cloud profile made by the aircraft, while each row represents a different measurements. The second, third, fourth and fifth rows are equivalent to the first, second, third and fourth rows of Figure 4.5. The additional row in this figure compares the temperature in the model to the temperature as measured by the Rosemount probe.

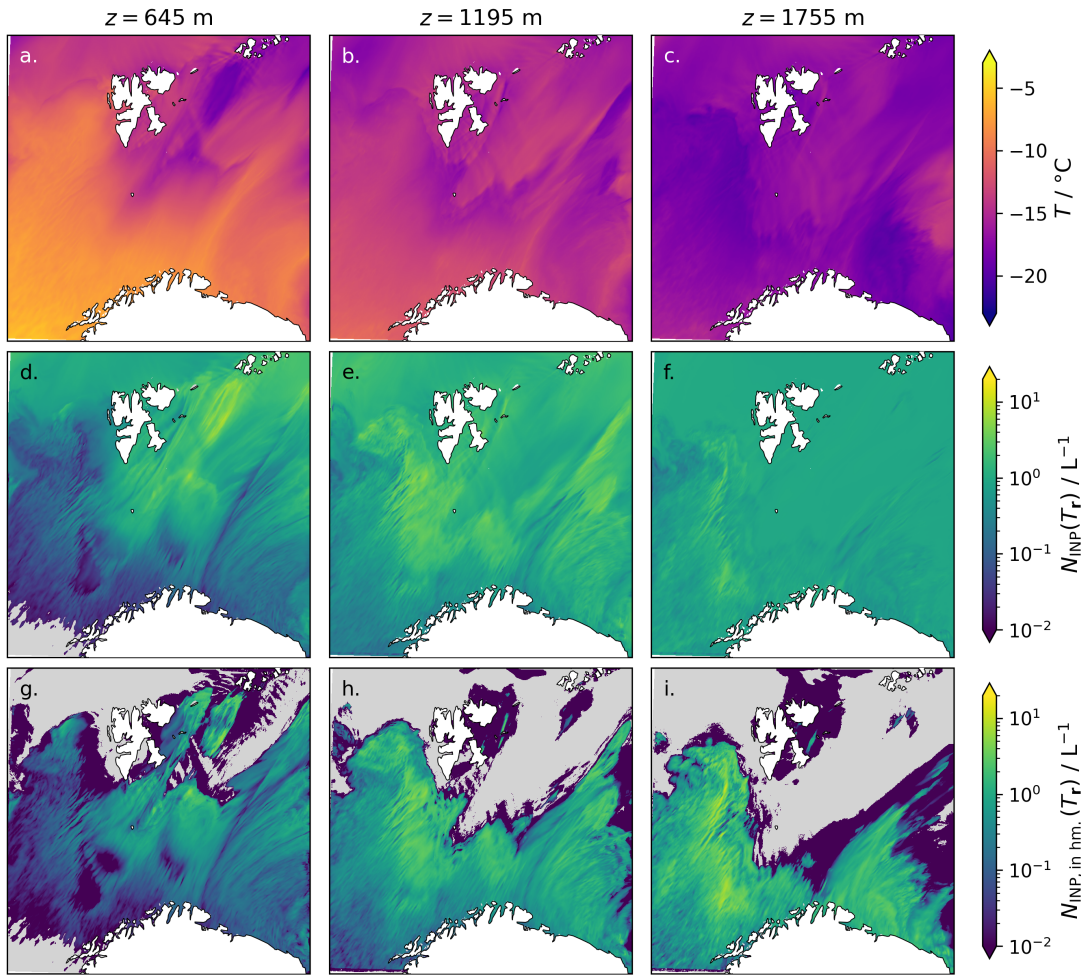


Figure C.3: Map of INP concentrations at three cloud-relevant heights, with INP concentrations shown at the temperature of the grid cell. Panels a, b and c show the temperature at heights $z = 645$ m, 1195 m and 1755 m respectively. The concentration of INPs at each grid point, $N_{\text{INP}}(T_r)$, is shown on these model levels in panels d, e and f, while the concentration of INPs contained within hydrometeors, $N_{\text{INP, in hm.}}(T_r)$ is shown in panels g, h and i. Areas where $T > -6$ °C or outside of cloud are shown in grey.

Appendix D

Appendices to Chapter 5

D.1 Profiles of INP concentration

Figure D.1 shows profiles of INP concentration in each analysis box with varying horizontal axes limits to enable easy comparison between each analysis box.

D.2 Transects of cold-air outbreaks

Figure D.2 shows the development of cloud through the CAO in each of the seven simulations.

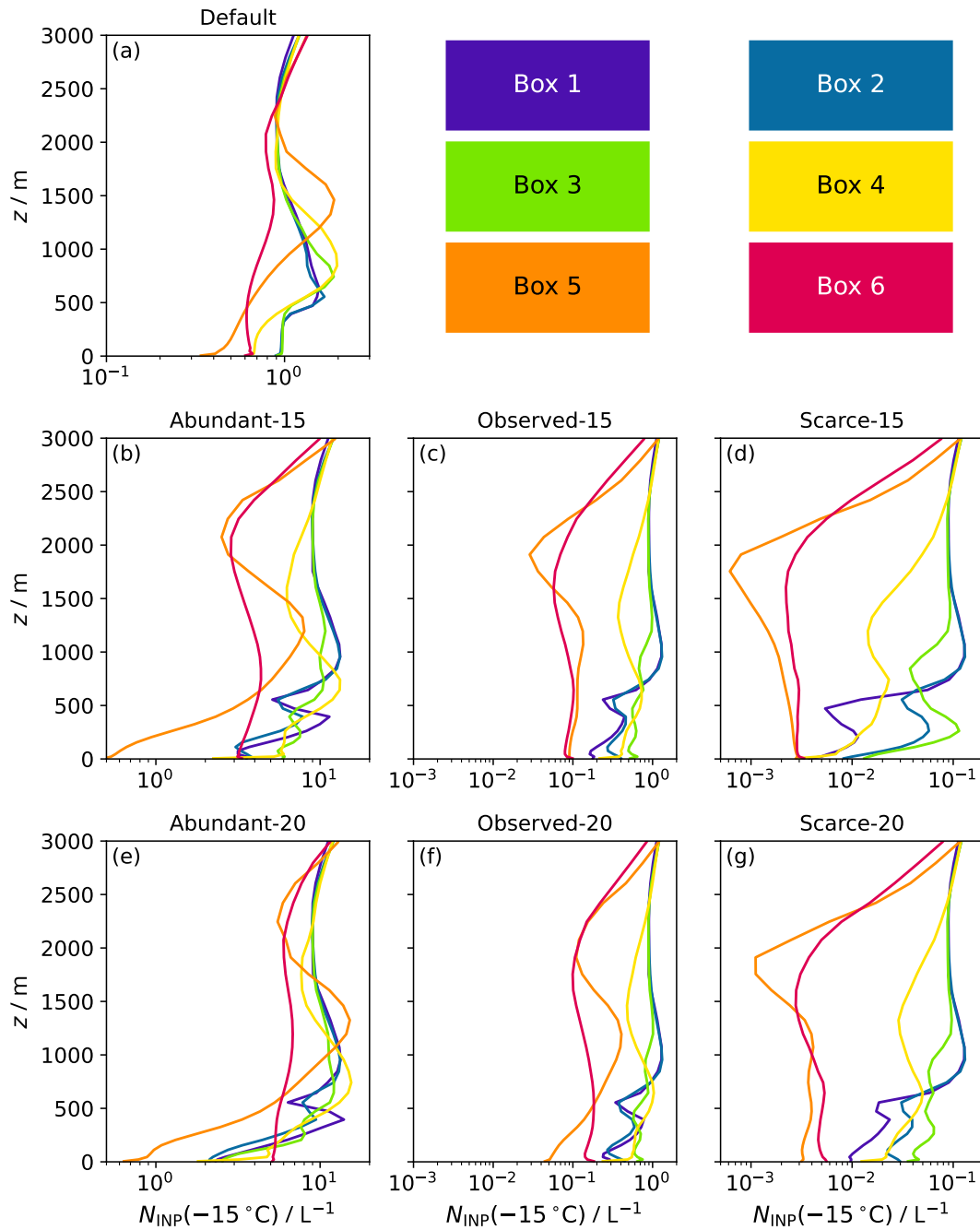


Figure D.1: Profiles of INP concentration in each of the six analysis boxes shown in Figure 4.3 for each of the seven simulations. As Figure 5.6, except horizontal axes limits differ between figures and have reduced scale to enable easier comparison of profiles within a simulation, rather than between figures.

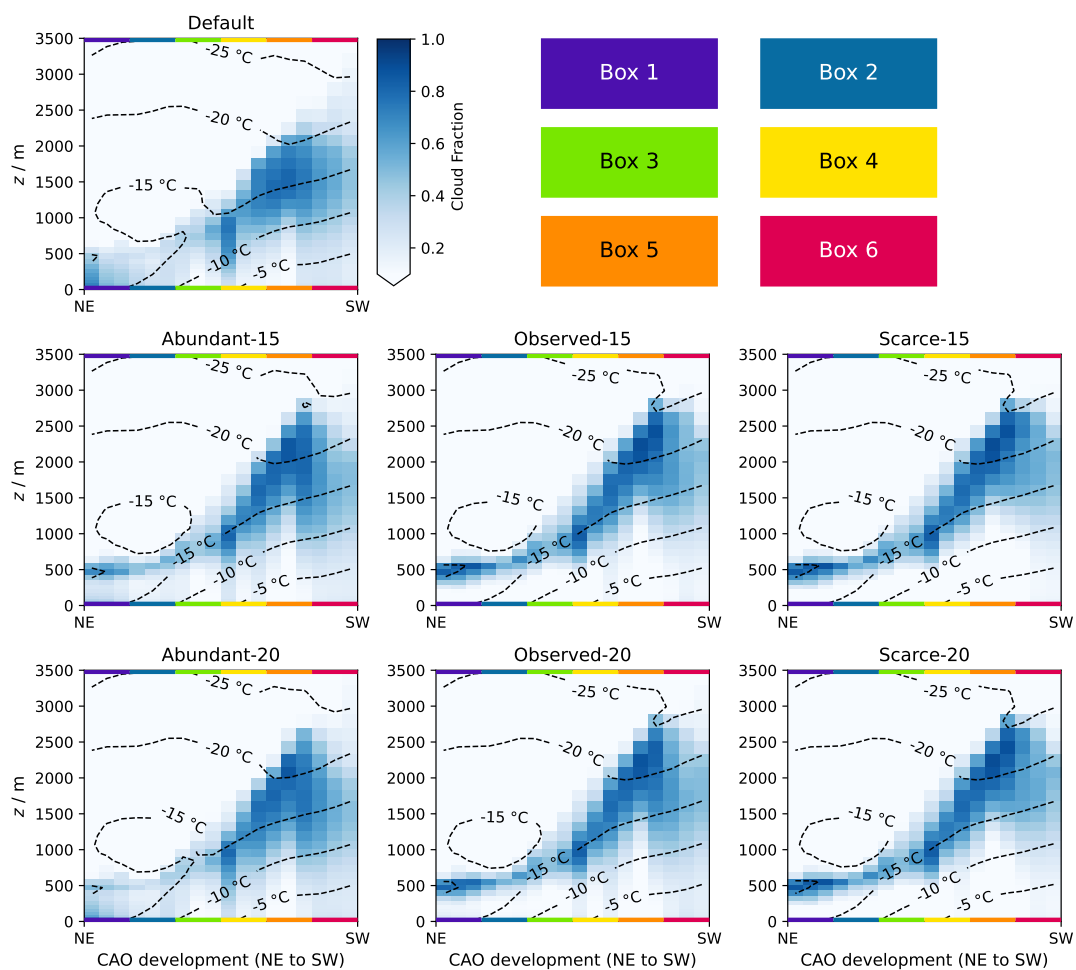


Figure D.2: Grid-box cloud fraction with temperature contours overlaid for all simulations.

Appendix E

Data availability

Raw and processed INP data and metadata, processed aerosol size data and INP data normalised by aerosol properties, HYSPLIT inputs and outputs, and Nevzorov clear-air flags can be found at:

<https://doi.org/10.5281/zenodo.11221599>.

Flight data from the FAAM aircraft are stored in the CEDA archive at:

<https://catalogue.ceda.ac.uk/uuid/01021a90c0c2481c909bdb145cb72398>

A list of UM suites used for modelling is provided in Table E.1. Code is proprietary, but namelists can be accessed via the Met Office Science Repository Service. Data for these suites is stored on the Met Office Managed Archive Storage System and can be accessed with the permission of the Met Office.

Table E.1: Suites used for modelling in Chapters 4 and 5

Simulation	UM suite
Default	u-du646
Abundant-15	u-du529
Observed-15	u-du527
Scarce-15	u-du530
Abundant-20	u-dw413
Observed-20	u-du528
Scarce-20	u-dw414

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