

A Review of the Factors Influencing Arctic Mixed-Phase Clouds: Progress and Outlook

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Abstract

Mixed-phase clouds are ubiquitous in the Arctic and play a critical role in Earth's energy budget at the surface and top of the atmosphere. These clouds typically occupy the lower and midlevel troposphere and are composed of purely supercooled liquid droplets or mixtures of supercooled liquid water droplets and ice crystals. Here, we review progress in our understanding of the factors that control the formation and dissipation of Arctic mixed-phase clouds, including the thermodynamic structure of the lower troposphere, warm and moist air intrusions into the Arctic, large-scale subsidence and aerosol particles. We then provide a brief survey of numerous Arctic field campaigns that targeted local cloud-controlling factors and follow this with specific examples of how the Arctic Cloud Observations Using airborne measurements during polar Day (ACLOUD)/ Physical feedback of Arctic PBL, Sea ice, Cloud And Aerosol (PASCAL) and Airborne measurements of radiative and turbulent FLUXes of energy and momentum in the Arctic boundary layer (AFLUX) field campaigns that took place in the vicinity of Svalbard in 2019 were able to advance our understanding on this topic to demonstrate the value of field

campaigns. Finally, we conclude with a discussion of the outlook of future research in the study of Arctic cloud-controlling factors and provide several recommendations for the observational and modelling community to advance our understanding of the role of Arctic mixed-phase clouds in a rapidly changing climate.

1 Introduction

The Earth's Arctic is warming at approximately twice the pace of the rest of the globe. This phenomenon, commonly known as Arctic amplification, is most pronounced during the late autumn and early winter (Serreze et al. 2009). Arctic amplification has been considered in various ways; previous studies have defined it as the ratio of warming in the Arctic to either global or tropical warming (Pithan & Mauritsen 2014, Stuecker et al. 2018, Middlemas et al. 2020), where the Arctic has been defined to be poleward of latitudes ranging from 60 to 75 °N and different definitions of the ratio have been used (Hind et al. 2016). Furthermore, the timescale of Arctic amplification has also been studied on both transient and equilibrium timescales (Holland & Bitz 2003, Tan & Storelvmo 2019). Here, we use Arctic amplification as an umbrella term that encompasses amplified warming in the Arctic relative to either the tropics or the entire globe and on either transient or equilibrium timescales (Yoshimori et al. 2016).

A number of mechanisms have been proposed to explain Arctic amplification (Serreze & Barry 2011, Taylor et al. 2013, Wendisch et al. 2017). An early mechanism proposed over half a century ago is the surface-albedo feedback, whereby warming induces the melting of snow and sea ice, which in turn induces further warming by reducing surface albedo (Budyko 1969, Sellers 1969). This positive feedback is among many others that have since been proposed. These include feedbacks related to the strength of surface temperature inversions (Boe' et al. 2009, Bintanja et al. 2011), poleward energy transport (Hwang et al. 2011, Merlis & Henry 2018, Graversen & Langen 2019) and clouds (Vavrus 2004, Cronin & Tziperman 2015, Tan & Storelvmo 2019, Wendisch et al. 2019, Middlemas et al. 2020) that may also interact nonlinearly with the surface-albedo feedback to further amplify or dampen Arctic warming. Several studies suggest that the sea-ice albedo feedback is the leading contributor to Arctic amplification (Manabe & Wetherald 1975, Hall 2004, Dai et al. 2019). Furthermore, it has been shown that the sea ice minimum in summer and early fall both directly and indirectly contributes to Arctic amplification through surface heat flux exchange (Screen & Simmonds 2010). However, simulations have indicated that Arctic amplification still occurs even when the surface-albedo feedback is locked (Graversen & Wang 2009), and a combination of energy balance and coupled climate models suggest that although the surface-albedo feedback plays a contributing role to Arctic amplification, it does not play a dominating role in climate models (Winton 2006, Pithan & Mauritsen 2014).

This review focuses on the processes and factors that control the evolution and properties of Arctic mixed-phase clouds known to influence Arctic amplification. These clouds consist of a combination of liquid droplets and ice crystals within the temperature range extending from 0 °C to the homogeneous freezing temperature of approximately -38 °C (Korolev et al. 2017). Although mixed-phase clouds are the focus of this manuscript, we note that some of the processes and factors considered herein also influence pure supercooled liquid clouds as well, especially in the summer where they are common in the Arctic boundary layer (Nomokonova et al. 2019). Since clouds were identified as the largest source of uncertainty in the climate

sensitivity in global climate models (Cess et al. 1989) decades ago, cloud feedbacks, i.e. the response of clouds to changes in surface temperature warming, continue to remain the largest contributor to uncertainty in climate projections today (Zelinka et al. 2020). However, substantial progress in narrowing the uncertainty range and clarifying the dominant mechanisms involved has been made throughout the decades (Zelinka et al. 2017), in large part due to improvements to model parameterizations and innovative diagnostic techniques such as satellite simulators that enable consistent comparisons between satellite observations and large-scale models (Bodas-Salcedo et al. 2011). The Arctic exhibits the largest spread in near-surface air temperature projections in climate models across all latitudes (Boucher et al. 2013). Arctic clouds are also especially poorly represented in climate models (Klein et al. 2009, Karlsson & Svensson 2011), with several models and even observations disagreeing on the annual cycle of cloud fraction (Boeke & Taylor 2016, Taylor et al. 2019). Although the magnitude of the contribution of the surface temperature-mediated cloud feedback to Arctic amplification was shown to be relatively small in the previous generation of climate models (Pithan & Mauritsen 2014), poor observational constraints on Arctic clouds combined with linear diagnostic techniques for highly nonlinear cloud feedbacks in the Arctic (Zhu et al. 2019) call into question the true sign and magnitude of the Arctic cloud feedback. Many of these low-level clouds are of the mixed-phase type that exhibit a unique vertical structure (Curry et al. 2000) and commonly exist as multilayer clouds, especially during the summer months. Due to the gradient in saturation vapour pressure over liquid and ice surfaces, these clouds are inherently unstable due to the Wegener-Bergeron-Findeisen (WBF) process (Korolev et al. 2003) and their maintenance and life cycle are difficult to represent in models of all scales (Korolev et al. 2017). In particular, climate models have a tendency to overestimate the proportion of ice crystals relative to the total cloud water in mixed-phase clouds (Komurcu et al. 2014, Cesana et al. 2015, McCoy et al. 2016), potentially due to the lack of a representation of subgrid-scale variability in cloud liquid and ice (Tan & Storelvmo 2016, Zhang et al. 2019) that is commonly observed in nature as revealed by aircraft in situ and remote sensing measurements (Korolev et al. 2003, Chylek & Borel 2004, D'Alessandro et al. 2019, Ruiz-Donoso et al. 2020). As such, climate models that parameterize the WBF process may be too active in climate models (Tan & Storelvmo 2016, Zhang et al. 2019), which may also lead to an excessive production of snow compared to observations (McIlhatten et al. 2017).

The thermodynamic phase of Arctic clouds and how it is distributed spatially is of critical importance for climate and radiation because the radiative properties of ice crystals and liquid droplets differ substantially; for the same water path and solar zenith angle, the reflectivity of water clouds can be up to four times greater than that of ice clouds (Sun & Shine 1994). This is due to the greater abundance and smaller size of liquid droplets relative to their solid counterpart, which is facilitated through the amount of cloud condensation nuclei (CCN) and ice-nucleating particles (INPs) available to initiate droplet and ice crystal formation in the atmosphere, respectively. Liquid clouds emit more downward longwave (LW) radiation to the surface compared to ice clouds. Here, LW radiation refers to radiation with wavelengths between 3-100 μm , where $\sim 99\%$ of Earth's outgoing LW radiation is emitted (Petty 2006). Solar radiation wavelengths, on the other hand, range from $\sim 0.25\text{-}4.0\mu\text{m}$, which includes the visible and shortwave infrared part of the electromagnetic spectrum. The LW radiation effect is particularly important during the polar night, however, it saturates when the cloud liquid water path reaches approximately 30 gm^{-2} (Shupe & Intrieri 2004), at which point clouds act as efficient blackbody

emitters, although the exact value for a given cloud depends on the average droplet effective radius (Wang et al. 2005). Downward LW radiation from clouds is also typically masked by increases in water vapour in a warmer climate, however, at constant relative humidity the LW cloud radiative effect (CRE), defined as the difference between all-sky and clear-sky cloud radiative forcing, was found to remain constant in the Arctic and, therefore, the sensitivity of the Arctic surface to LW CRE may increase in the future (Cox et al. 2015). Statistically significant multidecadal trends of cloud cover during spring and autumn based on data from ground stations in the Arctic Ocean also show increasing cloud cover that is conducive to sea-ice melt via surface warming from enhanced downward LW radiation (Eastman & Warren 2010). Ice crystals also tend to precipitate more efficiently as a result of their larger sizes, which affects cloud fraction and therefore the radiative impact of mixed-phase clouds. Therefore, the efficiency of the WBF process, which depends on how supercooled liquid and ice are spatially distributed within mixed-phase clouds, and in turn, the partitioning of the thermodynamic phase of mixed-phase clouds can therefore greatly impact the Earth's radiation budget and ultimately climate sensitivity (Mitchell et al. 1989, Tsushima et al. 2006, Tan et al. 2016, Frey & Kay 2018) and Arctic amplification as a result (Tan & Storelvmo, 2019).

A better process-level understanding of Arctic mixed-phase clouds is required to reduce the spread in the representation of these clouds in large-scale and high-resolution models. Here, we review a number of studies that have approached the problem using various observations, controlled experiments in large-eddy simulations and cloud-resolving models. We first provide an overview of Arctic mixed-phase cloud formation mechanisms (Section 2). This is followed by an up-to-date overview of the influence of the dominant factors that control Arctic clouds from the microscale to the synoptic scale, and from the inter-annual to the decadal timescale and beyond using multiple tools and observations in Section 3. These factors include temperature and moisture inversions, moisture intrusions, large-scale subsidence and aerosol particles. We emphasize the importance of taking into account the different surface types in the Arctic when considering the impact of the factors on clouds, classifying the interactions as belonging to either sea ice or open ocean categories. We argue that a comprehensive understanding of these factors is necessary to better constrain the impact of clouds on Arctic climate change and point out weaknesses that require more attention in future studies. Section 3 provides a broad overview of a number of Arctic field observations that studied the targeted the role of various cloud-controlling factors and demonstrates the utility of airborne and shipborne in situ observations. We conclude with a discussion of the outlook for future research on Arctic clouds and put forward several recommendations for improving our understanding of Arctic mixed-phase clouds in Section 4.

2 The formation and characteristics of Arctic mixed-phase clouds

Arctic mixed-phase clouds occur year-round with a minimum frequency of occurrence of 30% in the winter and 50% during the rest of the year based on spaceborne active remote sensing instruments limited to approximately 82°N (Mioche et al. 2015). Year-round ground-based remote sensing instruments in the Beaufort Sea observed mixed-phase clouds 41% of the time, with seasonal maxima in spring and autumn and cloud bases ranging from 0-2 km (Shupe et al. 2006). Their rather unique vertical structure often consists of a geometrically thin layer of supercooled liquid water droplets with a depth of approximately 0.5 km and a layer of ice virga

beneath it that commonly precipitates down to the surface (Curry et al. 1997, Shupe et al. 2006, de Boer et al. 2009, Verlinde et al. 2007, Mioche et al. 2017, Silber et al. 2020a). Liquid water-topped clouds are particularly common in the Arctic. Active satellite observations have shown that this distinct vertical structure of mixed-phase clouds is observed in 55 to 70% of all mixed-phase clouds in the entire Arctic domain, however with the caveat that these satellite observations inaccurately quantify the bottommost kilometre closest to the surface (Mioche et al. 2015). These clouds also occur at other latitudes; active spaceborne lidar and radar instruments observed their global frequency of occurrence to be ~8% based on (Zhang et al. 2010).

Advection of warm and moist air over the Arctic cold sea-ice surface initiates mixed-phase cloud formation and can lead to extensive stratus clouds (Herman & Goody 1976). LW radiation is then emitted to space from cloud-top (Pinto 1998), which decreases static stability and leads to the formation of eddies and a negatively buoyant overturning circulation (Nicholls 1984), analogous to the process occurring in subtropical stratocumulus clouds (Wood 2012). When supersaturation exceeds the equilibrium water vapour pressure of liquid water and ice surfaces, which is established through sufficient updraft velocities in the eddies, these turbulent eddies promote the growth of both thermodynamic phases --- liquid water and ice, rather than ice solely growing at the expense of the supercooled liquid droplets through the WBF process (Korolev 2007). The interactions of the cloud-top layer driven by radiative cooling with the surface below and/or the advected air aloft play a critical role in sustaining the liquid condensate and preventing the cloud from immediate glaciation (Solomon et al. 2018). Overall, a complex web of interactions between turbulent, radiative and dynamical processes collectively contribute to sustaining them, causing Arctic mixed-phase clouds to ultimately persist for days despite their inherent thermodynamic instability due to the WBF process (Morrison et al. 2012).

Multilayer clouds are also common in the Arctic, especially during summer (Curry et al. 1996, Shupe et al. 2011). These structures often consist of a low-level liquid or mixed-phase cloud and higher level mixed-phase or cirrus clouds (Vassel et al. 2019). These clouds usually form when large-scale horizontal advection associated with low and high pressure synoptic systems brings warm and moist air into the Arctic, which can condense in the presence of sufficient CCN and INPs at various levels. However, multiple cloud layers can also form within the lower troposphere (Verlinde et al. 2013), when sublimation of ice precipitation generated by the upper clouds results in cooling and moistening of the subcloud layer; this can lead to the formation of secondary inversion and a lower cloud (Harrington et al. 1999). The upper clouds of a multilayer system not only interact with radiation directly but also impact the shape, microphysical and radiative properties of the lower clouds. Overlying clouds shield lower layers from cloud-radiative cooling (Verlinde et al. 2013), which limits cloud-generated turbulent motions in the latter and thus condensation (Shupe et al. 2013). Moreover, ice precipitation from the higher layers into lower mixed-phase clouds can act as a sink for liquid water droplets through both vapour growth of the ice and rime collection (Verlinde et al. 2013). Enhanced riming can further reinforce secondary ice production and eventually trigger cloud glaciation (Lawson et al. 2015, Lloyd et al. 2015). Despite the critical impact of multilayer systems on the structure of the atmospheric radiative heating profile, the interactions of the individual layers are poorly quantified and understood. As a result, the microphysical structure of these systems is inadequately represented in models, which often overestimate (underestimate) the overall cloud liquid (ice) content (Morrison et al. 2009).

2 Factors that control Arctic clouds

We contend that the main factors that influence Arctic mixed-phase clouds include the thermodynamic structure of the Arctic atmosphere, which is determined by the frequent presence of temperature and moisture inversions, the oscillation of moisture intrusions that bring large bouts of moisture to the Arctic and the presence of CCN and INPs. Although large-scale subsidence is an important factor controlling subtropical clouds (Myers & Norris, 2013), evidence suggests that large-scale subsidence plays a secondary role in controlling the evolution of Arctic mixed-phase clouds. A discussion of each of these cloud-controlling factors and the interconnection between them follows.

2.1 Thermodynamic structure of the Arctic atmosphere

The vertical profiles of temperature and humidity define the thermodynamic structure of the atmosphere. The Arctic atmosphere's thermodynamic structure is strongly influenced by the frequent presence of low-level temperature inversions (Curry et al. 1996). In addition, the Arctic frequently experiences coincident specific humidity inversions (Devasthale et al. 2011). The degree to which temperature and moisture inversions impact Arctic cloud properties strongly depends on surface type, i.e. the extent to which the underlying surface is covered by sea ice, open ocean or land, whether the cloud layer interacts with the surface layer, and the degree to which the clouds are coupled to the surface (Sections 4.1.1 and 4.1.2). Arctic clouds are deemed "coupled" to the Arctic surface when cloud-driven turbulence interacts with surface turbulence (Figure 1d) and "decoupled" when the subcloud mixed layer remains disconnected from the

surface layer (Figure 1b and c). Field measurements indicate that decoupled clouds dominate both in winter (Gierens et al. 2020) and summer (Shupe et al. 2013, Sotiropoulou et al. 2014). Cloud-surface interactions can have important implications for a cloud's life-cycle (Shupe et al. 2013), especially from late spring to early October, when both ice-free ocean and melting sea-ice can supply clouds with enhanced moisture (Pinto & Curry 1995). Along with thermodynamic effects, clouds coupled to the surface may also be more affected by local sources of CCN (Mahmood et al. 2019) and INPs (Creamean et al. 2019), which may result in additional interactions and impacts on the Arctic's surface radiative energy budget and hydrological cycle (Section 4.4).

2.2 Temperature inversions

Temperature soundings from the year-long Surface Heat Budget of the Arctic Ocean (SHEBA) field campaign that took place in the Beaufort Sea in 1997 (Uttal et al. 2002) first revealed two preferred thermodynamic states over sea ice and snow-covered surfaces during the Arctic winter (Stramler et al. 2011). The most prevalent preferred state is characterized by strong surface inversions and cloud-free conditions or optically-thin ice clouds (Figure 1a) and is referred to as the "clear state". The second state is less preferred in the winter and dominates the summer and autumn seasons when clouds occur for 70-90% of the time and can persist for days to weeks (Shupe et al. 2011, Nomokonova et al. 2019, Zygmuntowska et al. 2012). The latter state is characterized by weaker, usually elevated temperature inversions (Tjernstroöm & Graversen 2009) and the formation of optically thick mixed-phase clouds (Figure 1b) and is referred to as the "cloudy state". Both "clear" and "cloudy" states have also been identified over open ocean and sea ice, such that altogether four states of typical atmospheric conditions in the ice-covered and open-ocean Arctic prevail (Wendisch et al. 2019).

The clear state is characterized by large net upward surface LW radiative energy flux densities of at least 40 Wm^{-2} . Downward LW emission from clouds in the cloudy state tends to offset surface cooling (Zuidema et al. 2005), often resulting in near-zero surface net LW radiation (Stramler et al. 2011, Graham et al. 2017). While the accurate representation of these two preferred states is critical for the correct representation of the Arctic surface radiative energy budget, climate models partly fail to reproduce the cloudy state and this causes systematically cold surface temperature biases and a stronger winter surface inversion (Pithan et al. 2014).

Satellite remote sensing and radiosonde observations show that temperature inversions tend to be stronger and occur closer to the surface over sea ice than open ocean (Pavelsky et al. 2011, Nygård et al. 2014). Although the temperature inversions are typically stronger during the winter months (Ganeshan & Wu 2015), averaging over 5 K, tend to be weaker during summer, averaging approximately 2K (Devasthale et al. 2011). The strength of the temperature inversions, as commonly quantified by the lower tropospheric stability (LTS), exerts a strong influence on Arctic low-level cloud properties, which in turn depends on whether the clouds are coupled or decoupled to the surface. Whether clouds are decoupled (Figure 1 c) or coupled (Figure 1 d) from the surface in turn is strongly influenced by surface type (Kay & Gettelman 2009). Over sea ice, atmospheric columns with stronger LTS tend to have fewer clouds, less cloud liquid water content, more cloud ice water content, and are also closer to the surface with a tendency for more frequent multi-layer clouds (Taylor et al. 2015, Taylor et al. 2019). Here,

clouds tend to be decoupled from the surface since it is generally colder than the overlying atmospheric boundary layer, which is influenced by the episodic advection of warm air from lower latitudes. Decoupling of the clouds from the surface leaves radiation exchanges as the only coupling mechanism. Previous studies found that climate models tend to overestimate the decoupling and the related Arctic LTS relative to observations (Medeiros et al. 2011, Barton et al. 2014). This was due to cold surface temperature biases over sea ice that arise from a lack of cloud liquid water path (Barton et al. 2014). On the other hand, a study using active satellite remote sensing observations showed that the opposite correlation holds over the ocean --- stronger LTS favours more cloud cover with more liquid water and less ice (Yu et al. 2019). Over areas of open ocean, that are common during autumn, warmer surface temperatures relative to the atmosphere enhance surface turbulent fluxes to the atmosphere and couple the clouds more often to the surface. This warming and moistening of the lower atmosphere can promote larger cloud fraction and liquid water content, and strong LTS can restrict boundary layer deepening and enhance low-level liquid clouds. The opposite effect occurs in the presence of weak LTS that reduces low-level liquid clouds over a coupled boundary layer through enhanced entrainment effects that are well-studied in subtropical marine stratocumulus clouds (Klein & Hartmann 1993, Wood 2012). For example, sensible heat fluxes that deepen the boundary layer were found to reduce cloud cover above Arctic wintertime leads (Li et al. 2020). A notable exception to the coupling of Arctic clouds to the surface over partially open ocean occurs during the summer, when clouds are decoupled from the surface due to a lag in sea ice melting that keeps the surface cooler than the overlying atmosphere. In general, over both surface types, stronger LTS promotes lower cloud bases. Moreover, in addition to the direct impact of LTS on cloud macrophysical properties, it can also indirectly influence aerosol-cloud interactions through the entrainment, transport and recycling of aerosol particles (Section 2.6).

2.3 Moisture inversions

Specific humidity or “moisture” inversions are often observed to accompany temperature inversions in humidity soundings and satellite profiles of water vapour at multiple layers in the Arctic atmosphere (Devasthale et al. 2011, Nygård et al. 2014). Over Arctic land, moisture inversions coincide with temperature inversions roughly 50% of the time on average. These moisture inversions form via two mechanisms: (i) surface or cloud-top radiative cooling and (ii) large-scale moisture transport (Naakka et al. 2018). In the former mechanism, LW radiative cooling at either cloud-top or at the surface can decrease the local moisture supply by promoting the condensation of water vapour and simultaneously cool the cloud-top or surface, thereby forming coincident moisture and temperature inversions. In the latter mechanism, moisture inversions are formed via the advection of moist air over a dry air mass. Over mountainous regions with large slopes such those that exist in Greenland, katabatic winds are commonly responsible for moisture inversions (Vihma et al. 2011). Note that katabatic winds over Greenland and wind-induced mixing have the opposite effect on the strength of moisture inversions; although the latter typically tends to erode moisture inversions, the former strengthens them.

The frequency, strength and mechanism of formation of Arctic moisture inversions varies with season and with surface type. Arctic moisture inversions tend to be stronger during summer. Annually, their strength varies from 0.2 - 2 gkg⁻¹. However, moisture inversions are more frequent in the winter (Devasthale et al. 2011, Naakka et al. 2018), occurring up to 80% of the

time, with depths ranging from 200-900 m (Nygård et al. 2014, Sotiropoulou et al. 2016). While the surface radiative cooling mechanism controls their frequency in the winter, the moisture advection mechanism is considered the dominant formation mechanism in summer (Naakka et al. 2018) when the Arctic Ocean is frequently affected by the transport of southerly warm and moist air (Naakka et al. 2018, Tjernström et al. 2019), although episodes of intense poleward moisture transport, known as moisture intrusions (Section 4.2) have been reported year-round (Pithan et al. 2018). Over sea ice, the frequency of co-existent moisture and temperature inversions is higher (Sedlar et al. 2012, Sotiropoulou et al. 2016) due to the fact that surface sensible and latent heat fluxes are generally small and of similar magnitude, which limits the instantaneous differences in boundary layer heat and moisture transport (Nygård et al. 2014).

Although moisture inversions are not unique to the polar regions, they do occur much more frequently in the polar regions relative to warmer latitudes and they are also structurally different in the Arctic compared to other regions. The increasing moisture supplies above the temperature inversion base often promotes condensation within the stable layer, allowing Arctic low-level clouds to extend into the inversion (Figure 1 b and c). This unique feature of Arctic clouds is commonly found over sea-ice (Sedlar et al. 2012) but not over open-water (Sotiropoulou et al. 2016). Moreover, the extension of the liquid layer into the inversion alters the effective cloud emission temperature; although it has a weak positive impact on LW irradiances at the surface ($\sim 1.5 \text{ Wm}^{-2}$) the increase in outgoing LW radiation at the top of the atmosphere can be up to 10 Wm^{-2} (Sedlar et al. 2012).

If the cloud layer is decoupled from the surface, elevated moisture inversions in the Arctic can play an important role in sustaining the lifetime of liquid clouds by entraining moist air from the inversion into the cloud and therefore increasing their optical thickness and radiative properties (Egerer et al. 2021). Idealized large-eddy simulations have shown that elevated moisture inversions can serve as a sufficient moisture source to maintain a decoupled cloud for days to a week, although this resulted in small differences in the properties of liquid and ice within a single mixed-phase cloud layer compared to the case with a coupled cloud without an elevated moisture inversion (Solomon et al. 2014). Thus, the existence of elevated moisture inversions implies that surface coupling is not the only source of moisture for Arctic clouds and that the cloud layer can evolve independently of whether it is coupled to the surface and is independent of the surface type. There is currently no consensus on the impact of surface coupling as a moisture source to maintain Arctic mixed-phase clouds based on the analysis of field observations; while some observational studies suggest that additional surface moisture sources enhance liquid condensate (Shupe et al. 2013, Gierens et al. 2020) others do not find a significant impact (Sotiropoulou et al. 2014). However, footprint-level satellite observations have shown that the influence of surface coupling on the evolution of cloud properties depends on the local atmospheric meteorological regime partitioned by LTS and mid-tropospheric vertical velocity (Taylor et al. 2015). The situation contrasts with cloud-surface decoupling in the mid-latitudes where decoupling promotes cloud break-up (Wood, 2012). Entrained air at cloud-top is usually dry air over the mid-latitude counterpart (Figure 1e) and thus cloud-top entrainment, evaporation and precipitation leads to cloud dissipation when the cloud system is disconnected from the surface vapour supply.

2.4 Warm and moist air intrusions

Several recent studies indicate that anomalously large moisture and heat transport from the south into the Arctic plays a critical role in Arctic amplification (Kapschetal. 2013, Woods & Caballero 2016, Johanssonetal. 2017, Messori et al. 2018). Such episodes bring warm and moist air into the Arctic and are often linked to extreme surface and sea-ice melting (Woods et al. 2013, Tjernstro'm et al. 2015, Park et al. 2015a, Park et al. 2015b, Park et al. 2015c, Boisvert et al. 2015, Hegyi & Taylor 2018). This phenomenon is sometimes referred to as warm and moist air intrusions or simply "moisture intrusions".

Wintertime moisture intrusions are supported by a synoptic blocking pattern to the east of the region affected (Woods et al. 2013). Moreover, a blocking system over the Ural Mountains can induce significant sea-ice decline in the Barents and Kara Seas when combined with the positive phase of the North Atlantic Oscillation, as this circulation pattern favours southerly moisture transport into the basins (Gong & Luo 2017). Several studies have linked the transport of enhanced moisture to poleward-propagating planetary-scale Rossby waves triggered by tropical convection (Yoo et al. 2012, Lee 2014, Park et al. 2015c, Baggett et al. 2016), which is referred to as the "tropically excited Arctic warming mechanism".

Moisture intrusions exert a substantial influence on Arctic cloud conditions. When transported air masses originate from open-water, they cool and condense when advected over sea-ice, resulting in cloud formation (Ali & Pithan 2020). Moisture intrusions are also one of the large-scale moisture transport mechanisms that lead to moisture inversions described in Section 4.1.2 that sustain clouds that are decoupled from the surface. Many studies have linked the occurrence

of moisture intrusions with increased cloudiness (Persson et al. 2017, Johansson et al. 2017, Liu et al. 2018, Messori et al. 2018) and enhanced downward LW radiation (Woods et al. 2013, Park et al. 2015a, Park et al. 2015b, Park et al. 2015c, Messori et al. 2018) and thus surface warming. Large-eddy simulations reveal that advected heat has a relatively weak impact on cloud properties whereas moisture is crucial for cloud maintenance in the Arctic boundary layer (Sotiropoulou et al. 2018). In contrast, the advection of warm and moist air within oceanic boundary layers is associated with a negative impact on cloud fraction (Knudsen et al. 2018, Eirund et al. 2019). Heat and moisture increase within the boundary layer promote destabilization and convection in the lower atmosphere (Eirund et al. 2019). As a result, the stratocumulus cloud breaks up, the cloud-top is lifted and cloud fraction is substantially reduced. Decreases in cloud fraction and cloud liquid water content result in enhanced outgoing LW radiation and surface cooling. However, when advection occurs above the boundary layer, it can promote the formation of multilayer structures and thus an overall increase in liquid water path (Eirund et al. 2019).

While infrequent, moisture intrusions are responsible for the bulk of the poleward moisture transport in the Arctic during both winter and summer (Liu & Barnes 2015). Moreover, the associated anomalies in moisture content and cloudiness have been linked to accelerated onset of the sea-ice melting period (Kapsch et al. 2013, Mortin et al. 2016). Yet, despite their climatic significance (Pithan et al. 2018), moisture intrusions are poorly represented in climate models, which fail to reproduce their regional characteristics (Woods et al. 2017). These deficiencies can have a substantial impact on Arctic cloud representation in climate models, as models with enhanced poleward heat and moisture transport produce improved cloud fractions and cloud liquid properties (Baek et al. 2020). Resolving these model issues requires dedicated measurement campaigns in a Lagrangian air parcel framework (Pithan et al. 2018, Wendisch et al. 2021).

2.5 Large-scale subsidence

While several recent studies have focused on the impact of large-scale advection on Arctic clouds, less is known about the impact of subsidence, which often accompanies poleward atmospheric transport (Tjernström et al. 2019, Neggers et al. 2019). Large-scale subsidence is weaker in the Arctic compared to the subtropics and thus potentially plays a lesser role in the Arctic. Dedicated measurements of synoptic-scale divergence and derived vertical pressure velocity, such as those performed in the subtropics (Stevens et al., 2021) are scarce in the Arctic; however, there are plans to conduct appropriate samplings (Wendisch et al. 2021). In the Arctic, large-scale subsidence is weak and can be thought of as being correlated with the generation of a surface temperature inversion --- as the air is advected over the central Arctic from lower latitudes it slowly sinks as the air radiatively cools (Tjernström et al. 2019). Thus, unlike the case in the subtropics, subsidence is not an active driver of the inversion strength and is merely correlated with stronger LTS and inversion strength. In this section, we discuss our current limited knowledge of the role of subsidence in Arctic clouds based on a limited set of field observations, large-eddy simulations, as well as large-scale climate models and satellite observations.

Reanalysis data and field observations from the Arctic Clouds in Summer Experiment (ACSE) have revealed that air parcels were higher in altitude and further south a few days before the presence of a surface inversion (Tjernström et al. 2019). Although subsidence is generally linked with the presence of a surface temperature inversion over melting sea-ice, moisture and cloud characteristics are more variable (Tjernström et al. 2019). In particular, the formation of moisture inversions and low clouds or fog was found to be associated with weaker subsidence, compared to cases where the stratified boundary layer is drier and often cloud-free. While reanalysis data suggests that subsidence weakly enhances the fraction and liquid water path of Arctic clouds (Zhao & Wang 2010), field observations suggest that subsidence is correlated with the existence of optically thinner Arctic clouds, potentially by impacting the entrainment of aerosol particles into the boundary layer (Zuidema et al. 2005).

The limited number of large-eddy simulations has shown contradictory results in terms of the role of subsidence in Arctic low-level cloud properties. In a study employing large-eddy simulations, increases in large-scale subsidence result in more turbulent clouds with enhanced liquid condensate over open-water (Young et al. 2018). The enhanced subsidence reinforces the boundary layer inversion strength and reduces entrainment of warmer air aloft. This allows for greater cloud liquid, thus more efficient precipitation, cloud-top radiative cooling and downdraft turbulent production. The combination of strong cloud-top radiative cooling, sub-cloud rain evaporative cooling and latent heat release from snow growth at cloud base destabilize the boundary layer, resulting in more convective structures (Young et al. 2018). However, other studies showed that enhanced subsidence resulted in an overall decrease in cloud liquid water content over sea-ice due to entrainment of drier air (Dimitrelos et al. 2020). In line with this study, strong and sudden subsidence led to cloud dissipation when the boundary layer top was pushed below the lifting condensation level (Neggers et al. 2019). The cloud response to variations in large-scale vertical forcing for different surface, thermodynamic and microphysical conditions has not been comprehensively explored. Additional studies with cloud resolving simulations, preferably in a Lagrangian framework (Neggers et al. 2019, Dimitrelos et al. 2020), are necessary to improve the understanding of the role of subsidence in Arctic cloud evolution and radiation (Wendisch et al. 2021).

Finally, the role of subsidence as an Arctic cloud-controlling factor is also limited to a few studies employing large-scale climate models and satellite observations. Synergistic CloudSat/CALIPSO observations and reanalysis-based regimes have shown that the Arctic atmosphere produces a wide range of 500 hPa vertical pressure velocity values (ω_{500}) ranging from weak ascent to strong sinking motion for each of the LTS-based regimes (Barton et al. 2012, Taylor et al. 2015). Under conditions of large-scale subsidence, the altitude of low-level clouds was very sensitive to LTS. Satellite observations and a climate model also show that stronger subsidence may also increase relative humidity in the lower troposphere (Curry et al. 1988), and in turn trigger Arctic sea-ice melt in the summer via enhanced downward LW cloud radiation at the surface (Huang et al. 2021). However, the dependence of cloud properties on ω_{500} was shown to be much weaker than that on LTS in the CMIP5 models (Taylor et al. 2019), suggesting a relatively minor role compared to other cloud-controlling factors in the context of large spatial and temporal scales.

2.6 Aerosol particles

The influence of aerosol particles on the cloud microphysical properties that drive cloud radiative effects is poorly quantified yet is of fundamental importance to Earth's climate and its future change (Fan et al. 2016). Although Arctic low- and mid-level cloud properties and radiative effects can be highly susceptible to aerosol effects on a local scale (Garrett & Zhao, 2006, Lubin & Vogelmann, 2006), regional-scale impacts have not been thoroughly explored. A series of recent reviews on topics related to Arctic aerosol distributions, mixed phase cloud modeling, microphysics, and aerosol interactions show that regional uncertainty in aerosol-cloud interactions (ACIs) is in large part due to (i) CCN and INP levels being difficult to predict, and (ii) aerosol impacts on clouds being microphysically complex and linked to meteorology (Morrison et al. 2012, Fan et al. 2016, Kanji et al. 2017, Lohmann 2017, Fridlind et al. 2007, Willis et al. 2018, Schmale et al. 2021). This section complements these reviews by linking the larger-scale meteorological influences discussed in the sections above with what is known about Arctic-specific aerosol sources and microphysical cloud impacts. Specifically, we discuss the factors affecting the concentrations and activity of Arctic CCN and INPs, the robust and uncertain mechanisms by which these CCN and INPs impact radiation-relevant cloud properties, and how aerosol cloud interactions may be changing with a warming Arctic.

2.6.1 Sources, concentrations, and activity of Arctic CCN and INPs

CCN and INPs are derived from marine, terrestrial, and anthropogenic sources. Combustion-derived aerosols are sporadically transported to the Arctic from lower latitudes (Soja et al. 2008) and there are local near-surface aerosol particle sources from exposed glacial till dust (Zwaafink et al. 2016, Tobo et al. 2019), shipping and oil extraction (Schmale et al. 2018), and thawing permafrost (Creamean et al. 2020). However, more commonly, summertime Arctic aerosol particles are produced from local marine primary and secondary sources. These sources can provide at least half of the CCN supply to the Arctic atmosphere via primary sea spray emissions (Quinn et al. 2017) and new particle formation (Dunne et al. 2016, Heintzenberg et al. 2017, Merikanto et al. 2009, Yu & Luo 2009). Marine aerosols may also supply more than half of all INPs at high latitudes (Burrows et al. 2013, Wilson et al. 2015). In contrast, during winter and

spring, marine biogenic aerosol particle concentrations are lower when the open ocean is less exposed to the atmosphere due to both greater sea ice cover and lower biological productivity, and aerosol particles derived from long-range transport, dust and combustion sources are more prevalent (Barrie 1986, Stohl 2006, Quinn et al. 2007, Arrigo 2008, Engvall et al. 2008, Croft et al. 2016), although marine sources of INPs are still present to some extent (Hartmann et al. 2019b). Sea salt aerosol particle concentrations can be larger in winter, potentially due to upward migration of brine from underlying sea ice and subsequently lifting and sublimation in the atmosphere through a strong influence from blowing snow (Huang & Jaegle 2017). Long-range transport of aerosol particles to the Arctic from sources such as dust and smoke (Bullard et al. 2016) as well as biomass burning aerosols derived from boreal forest fires especially during the spring season (Marelle et al. 2015) are also important sources of Arctic CCN and INPs. Long-range transported wintertime aerosols can accumulate and form Arctic haze due to the combination of a cold, stable boundary layer and reduced particle and gas removal rates (Shaw 1995). In particular, black carbon is an important contributor of Arctic haze and its wintertime peak is controlled by its hydrophilic fraction and weak wet deposition rate (Shen et al. 2017).

Concentrations of CCN typically range between 1-100 cm⁻³ at supersaturations between 0.3-0.8%, but “tenuous” regimes with CCN concentrations < 10 cm⁻³ have been observed during multiple field campaigns (Bigg & Leck 2001, Bigg et al. 1996, Lannefors et al. 1983, Leaitch et al. 2016, Leck & Svensson 2015, Leck et al. 2002, Mauritsen et al. 2011, Tjernstro m et al. 2014). CCN levels are lowest during the summer, when midlatitude aerosol transport is inefficient and midlatitude wet deposition is likely to scavenge long-range transported aerosols before they reach the Arctic (Bourgeois & Bey 2011, Di Pierro et al. 2013, Law & Stohl 2007, Quinn et al. 2007). Unlike CCN and aerosols in general, Arctic INPs appear to be more prevalent during the summer (Wex et al. 2019), although overall their concentrations tend to be quite low, ranging from 10⁻⁴ – 10⁻² L⁻¹ at -15°C (Mason et al. 2016, Si et al. 2018, Creamean et al. 2019, Hartmann et al. 2019a, Irish et al. 2019, Wex et al. 2019), although concentrations have been observed as high as 0.25 L⁻¹ (Bigg 1996). Ice core data suggests that the summertime INP peak is caused by biological aerosols of marine and possibly terrestrial origin (Hartmann et al. 2019b). Ship-based CCN observations from (Wendisch et al. 2019) were lowest when their research vessel was surrounded by sea ice and highest during open ocean conditions, which supports the role of local marine emissions. However, in those locations where dust is present, dust may be an equal or more significant source of INPs than biogenic aerosols (Si et al. 2018, Vergara-Temprado et al. 2017, Abbatt et al. 2019, Irish et al. 2019, Huang et al. 2018, Tobo et al. 2019). Local dust is more exposed during the summer, but long-range transported dust may be more common in the winter.

The distributions and ability of CCN and INPs to impact Arctic clouds are quite heterogeneous in space and time (Willis et al. 2018, Moore et al. 2011), and are difficult to characterize because both CCN and INPs can be modified during atmospheric transport. For example, polluted aerosols are not thought to be major sources of INPs at the temperature ranges important to mixed phase clouds (Borys 1989, Hartmann et al. 2019b). Moreover, if they mix with INPs from other sources, the INP activity of these other particles may be reduced after being coated with sulphuric acid, and they may not freeze until colder temperatures (Girard & Asl 2014, Borys 1989, Cziczo et al. 2009, Eastwood et al. 2009, Grenier & Blanchet 2010, Tan et al. 2014, Coopman et al. 2018b). However, it is important to note that while some studies found that

pollution coatings can decrease the ice-nucleating ability of certain INPs, others found no impact (Archuleta et al. 2005, Knopf & Koop 2006). Given the generally low concentrations of aerosols in the region, the impact of pollution coatings may be particularly important for Arctic aerosol cloud interactions (Coopman et al. 2018a).

Neither CCN nor INPs can be accurately measured in the Arctic with current satellite remote sensors, so much of our information on their distributions relies on field data. Arctic INP field data are rare and are associated with non-negligible uncertainties (Demott et al. 2015, Garimella et al. 2018). The link between CCN and INP distributions and ambient meteorological conditions adds another level of challenge to CCN and INP prediction. At temperatures below $\sim -15^{\circ}\text{C}$ in the Arctic, INPs can be activated from sources that otherwise might not have been important at lower latitudes (Wilson et al. 2015, Kanji et al. 2017) where INPs cannot get lofted to high enough altitudes with sufficiently cold temperatures outside regions of deep convection. At the same time, Arctic INPs are able to nucleate ice at temperatures as high as -5°C (Wex et al. 2019), although activation at these warm temperatures are typical for INPs composed of bacteria that can also be found at other latitudes (Murray et al. 2012). Moreover, INPs become less effective at warmer temperatures, and more INPs and CCN become active at higher water vapour supersaturations. CCN are easier to sample than INPs, but their distribution is also not well known. For example, in very clean Arctic conditions, even particles as small as 20-30 nm can nucleate cloud droplets at high water vapour supersaturations (Burkart et al. 2017, Croft et al. 2016, Koike et al. 2019, Leaitch et al. 2016). These potentially cloud-active aerosol particles are too small to be detectable from current cooling, orne instruments (Hallen & Philbrick 2018), and must be sampled by in situ measurements. Moreover, the aforementioned tenuous aerosol layers consisting of low concentrations of CCN and INPs that are very optically thin are also not detectable from spaceborne instruments (Winker et al. 2013, Cho et al. 2013). For these reasons, CCN and INP distributions are least well-constrained outside of clouds, at high altitudes and over remote regions where few CCN and INP field data exist.

2.6.2 Aerosol-cloud-radiation interactions

Models consistently show that aerosol impacts in Arctic mixed-phase clouds are large enough to potentially affect sea ice melt (Jiang et al. 2000, Shindell & Faluvegi 2009, Gagné' et al. 2017, Regayre et al. 2015, Mahmood et al. 2019, Koch et al. 2009, Alterskjær et al. 2010, Dalsøren et al. 2013). For example, INP levels have a large impact on modelled cloud phase, cloud fraction and precipitation (Fridlind et al. 2012, Prenni et al. 2007, Ovchinnikov et al. 2014, Morrison et al. 2011), with potentially important impacts on surface CREs (Shupe & Intrieri 2004), top-of-the-atmosphere CREs (Xie et al. 2013, English et al. 2014), and ultimately, Arctic amplification (Tan & Storelvmo 2019). Enhanced INP levels will affect heterogeneous ice crystal formation and growth processes, for example via immersion freezing (de Boer et al. 2010). INP-driven glaciation could affect cloud lifetime and precipitation through either liquid removal in the cloud or through the WBF process referred to as the "glaciation effect" (Curry 1995, Lohmann 2002) and/or associated secondary ice multiplication (Field et al. 2017, Korolev & Leisner 2020). Conversely, deactivation of pre-existing INPs when pollution aerosols are present can reduce ice nuclei levels and glaciation (Girard et al. 2005, Lohmann 2017).

However, clear in situ evidence is still lacking for how often these processes occur or how important they might be in part because many past aircraft campaigns (Table 1) have missed key parameters, such as INP or CCN levels, aerosol composition, or ice cloud particle habit. Also, parameterizations for ice phase processes in mixed-phase clouds lead to large uncertainties in modelled cloud properties that have not yet been resolved (Tan & Storelvmo 2016, Boucher et al. 2013, Xie et al. 2013, Liu et al. 2011, Morrison et al. 2011, Klein et al. 2009, Taylor et al. 2019). Moreover, while INP-related processes appear to drive cloud microphysical and radiative responses to aerosols in some conditions and locations (Solomon et al. 2018, Costa et al. 2017, Jouan et al. 2012), CCN-related processes may be more important in other situations (Lance et al. 2011, Norgren et al. 2018). For example, the role of CCN-driven processes seems to be particularly important in tenuous mixed-phase cloud regimes with very low cloud droplet number concentrations (Loewe et al. 2017, Stevens et al. 2018).

There are also various other ways that CCN can impact mixed phase clouds. Enhanced levels of CCN lead to smaller and more numerous liquid cloud droplets in Arctic mixed-phase clouds on average over large spatial and time scales (Coopman et al. 2018a). Smaller droplets can affect cloud lifetime and precipitation by (i) impeding liquid droplet precipitation (Albrecht 1989), (ii) reducing liquid droplet collection from falling ice particles (Borys et al. 2000, Lohmann et al. 2003) known as the “riming indirect effect” (Lohmann 2017), and (iii) reducing secondary ice production from collision and splintering processes (Rosenfeld 2000). Cloud lifetime and droplet size in thin clouds affect cloud LW radiative emissivity (Shupe & Intrieri 2004), which in turn impacts moisture and surface turbulent fluxes, cloud-top cooling, and mixed layer depth (Solomon et al. 2018, Lubin & Vogelmann 2006, Garrett & Zhao 2006, Garrett et al. 2009). In summer, smaller and more numerous droplets at constant liquid water content will also cause more radiation to be scattered back to space (Twomey 1977). Multi-layer clouds are commonly observed in this region (Liu et al. 2012), and changes to mixed-phase cloud CCN-driven precipitation could also affect seeding of lower-level clouds. Subsequent seeding-related changes to cloud ice and precipitation formation (Luo et al. 2008, Silber et al. 2020a, Vassel et al. 2019) may then affect cloud dissipation and surface albedo. In some conditions both CCN and INPs might drive co-occurring processes that can behave nonlinearly.

Currently, the concentrations of CCN and INPs are a major source of uncertainties in models of Arctic mixed-phase clouds. These concentrations are particularly poorly constrained within clouds, where they can be entrained (Avramov et al. 2011, Igel et al. 2017), redistributed (Solomon et al. 2018), scavenged and precipitated (Morrison et al. 2005, Willis et al. 2018). INPs may also be sublimated and then re-entrained (Solomon et al. 2015, Possner et al. 2017, Verlinde et al. 2007, Fan et al. 2009) and they may become more efficient when CCN-related processes like LW cloud-top radiative cooling lower cloud temperatures and increase immersion freezing rates (Fu & Xue 2017, Possner et al. 2017). The uncertainty in aerosol impacts on cloud phase are another major issue in models. Cloud phase, along with cloud fraction, exerts a large influence on CREs. Aerosol impacts on cloud phase are poorly constrained in global climate models not solely in the Arctic but also globally (Karlsson & Svensson 2011, Cesana et al. 2015, Tan et al. 2016, McCoy et al. 2016, Taylor et al. 2019).

Field and remote sensing data offer complementary insights to models of aerosol interactions in Arctic mixed-phase clouds, but must be viewed in light of their own uncertainties. A main

challenge is that aerosols often co-vary with meteorological factors that control clouds as discussed in the previous sections. For example, across Arctic sea ice regions during polar night, satellite observations showed that combustion aerosols are associated with an average 10 Wm^{-2} difference in surface LW CREs, but that between 57-91% of this signal is caused by changes in meteorological conditions associated with aerosol transport (Zamora et al. 2018). Other challenges with interpreting field data include that ice concentrations within clouds are difficult to measure accurately (Fridlind et al. 2007), and microphysical processes can be impacted by co-occurring cold-weather phenomena, including secondary ice production (Field et al. 2017, Korolev & Leisner 2020) and seeding from either Arctic multi-layer clouds above the mixed-phase cloud layer (Luo et al. 2008) or frost flowers (Xu et al. 2016). Moreover, the sign, magnitude, and mechanisms by which aerosols impact Arctic mixed-phase cloud precipitation and radiative effects vary, depending not only on CCN and INP levels, but also on incoming radiation and surface albedo, multi-layer cloud radiative shielding, and cloud properties such as height, temperature, and liquid water content (Quinn et al. 2008, Shupe & Intrieri 2004, Sedlar et al. 2011, Willis et al. 2018, Sedlar & Shupe 2014, Morrison et al. 2012, Stofferahn & Boybeyi 2017). For example, at Utqiagvik, Alaska, aerosol microphysical effects on clouds may lead to surface heating as large as 12 Wm^{-2} in winter, and surface cooling as large as 12 Wm^{-2} in the summer (Zhao & Garrett 2015). Therefore, to quantify aerosol-cloud effects across the Arctic, observations are needed over large spatial and temporal scales, with attention paid to verifiably accounting for meteorological co-variability with aerosols.

When data are compared over large temporal and spatial scales and across cloud types, most remote sensing-based observations seem to agree that combustion and dust aerosols are associated with some combination of higher glaciation temperatures, more cloud ice, more precipitation, and reduced cloud fraction in the Arctic (Zhang et al. 2018, Coopman et al. 2018b, Filioglou et al. 2019, Zamora et al. 2018, Villanueva et al. 2020, Coopman et al. 2020). Aerosols have been associated with less cloud ice and less precipitation at specific locations or in specific cloud types (Zamora et al. 2017, Norgren et al. 2018), but these trends seem to be associated with combustion aerosols (Filioglou et al. 2019) and might be caused by either a CCN-related process or by a deactivation effect. Even presuming that the glaciation effect is dominant now (and more work is still needed to verify this hypothesis), it is unclear whether this process will remain dominant in a future warmer, wetter, and more aerosol-laden Arctic environment. Either way, for the reasons discussed above, aerosol-related uncertainties contribute to major uncertainties in climate projections (Bellouin et al. 2020), especially for the Arctic where rapid changes to historic aerosol, moisture, and heat fluxes are expected.

2.6.3 Aerosol-meteorology interactions and their impact on clouds and radiation

Aerosol impacts on radiation-relevant cloud properties have a strong relationship with meteorological conditions, such as temperature. Warmer temperatures reduce INP effectiveness and glaciation and riming processes (Eirund et al. 2019). As Arctic INPs are thought to have a large influence on mixed phase cloud processes (Section 4.4.2) this warming could become increasingly important to cloud dynamics. Warmer temperatures can also affect the microphysical environment in which aerosols are suspended, influencing the degree to which aerosols contribute to the Twomey effect, the WBF process, precipitation, splintering, and riming, and more generally, the potential importance of CCN compared to INP-dominated

aerosol microphysical processes. Thus, although many uncertainties remain, INP-driven glaciation processes might become less influential in the future warmer Arctic.

Besides temperature, other related meteorological parameters can influence Arctic ACIs as well, such as decoupling with the surface (Creamean et al. 2021), as well as stability and moisture levels. Besides affecting temperature, decoupling limits the influence of surface aerosol sources on clouds at higher altitudes, but promotes recycling of INPs that are released during sublimation of precipitating ice particles at the base of the subcloud layer (Fan et al. 2015, Solomon et al. 2018). This process, which plays a critical role in maintaining cloud-phase partitioning (Solomon et al. 2015, Solomon et al. 2018), is likely not favoured in coupled clouds (Kalesse et al. 2016). A more stable atmosphere (such as over sea ice) promotes weaker cloud-top entrainment of free-tropospheric air that can serve as moisture and a source of CCN and INP concentrations in the cloud layer (Solomon et al. 2011, Solomon et al. 2014, Morrison et al. 2012, Fridlind et al. 2007, Coopman et al. 2018b). It may also concentrate aerosols emitted from local sources (Willis et al. 2018). Atmospheric moisture content, which is impacted by temperatures, atmospheric stability and moisture intrusions, influences aerosol deposition and loss processes (Browse et al. 2012), and in turn impacts a CCN/INP's lifetime potential for impacting clouds. More moist and less stable conditions over open ocean can also activate smaller CCN particles, which might then affect cloud droplet feedbacks with mixed-phase cloud vertical mixing and radiative cooling (Silber et al. 2020b).

Moisture intrusions may also influence ACIs, particularly in areas decoupled from the surface. These intrusions are often aerosol-laden, and they produce not only in more aerosol transport (Thomas et al. 2019), but also in more frequent precipitation and aerosol loss. The extension of cloud top into the inversion layer modulates aerosol fluxes into the cloud as well as moisture entrainment fluxes and can thus impact cloud lifecycles (Solomon et al. 2011, Egerer et al. 2020, Igel et al. 2017). Other changes, for example in large-scale subsidence, might also affect the cloud microphysical environment upon which aerosols operate, for example impacting cloud-top radiative cooling rates and ice and liquid water paths, as well as aerosol entrainment rates and precipitation loss rates (Young et al. 2018, Brooks et al. 2017, Dimitrelos et al. 2020).

Multiple studies have found Arctic cloud responses to non-marine aerosols to be clearly reduced over open ocean compared to sea ice (Zamora et al. 2017, Zamora et al. 2018, Eirund et al. 2019, Filioglou 2019). That there would be a difference between open ocean and sea ice ACIs is not surprising, given that the two surface types produce very different levels of stability and aerosol, heat, and moisture fluxes and aerosol emissions (Wendisch et al. 2019, Willis et al. 2018, Schmale et al. 2021). For example, not only are marine aerosol levels much larger over the open ocean than over sea ice, but clouds over open ocean generally experience warmer and wetter conditions compared to those over sea ice. Aerosols may at times also impact meteorology, as when aerosol-driven increases in thin cloud LW radiative emissivity warm the surface and thereby increase moisture and heat fluxes (Morrison et al. 2012). Although the dominant causes for the observed differences between open ocean and sea ice are unknown, the trend of reduced aerosol influence over open ocean regions suggests that in the absence of significant new aerosol sources or pathways, the impacts of non-marine aerosols may become less influential in the future as the Arctic warms.

3 A brief survey of Arctic field campaigns targeting cloud-controlling factors

Despite limitations in their temporal and spatial coverage, in situ field observations are an indispensable tool for climate science by virtue of their relatively high accuracy and frequency of measurements relative to global satellite observations that can be used to validate regional models and develop model parameterizations. A number of field campaigns over the Arctic that took place mostly during the non-winter months have been performed over the past few decades (Figure 2a) and have been combined with ground-based stations in the Arctic (Figure 2b) to compensate for the limitations of spaceborne remote sensing. This section begins with a brief overview of a number of these field campaigns and some of the studies that have applied observations from them to gain insight on the influence of several factors that influence Arctic clouds. This is followed a short discussion of some lessons learned from three examples of the numerous field campaigns conducted in the past, namely the combined airborne and ship-based Arctic Cloud Observations Using airborne measurements during polar Day (ACLOUD), the Airborne measurements of radiative and turbulent FLUXes of energy and momentum in the Arctic boundary layer (AFLUX) and the Physical feedback of Arctic PBL, Sea ice, Cloud And Aerosol (PASCAL) field campaigns.

3.1 Overview

Several aspects of Arctic clouds, aerosols, radiation and their interactions were targeted by field campaigns. Selected examples that aimed to study the interaction of the thermodynamic and turbulent boundary layer structure with clouds include the Beaufort and Arctic Storms Experiment (BASE) (Gultepe et al. 2000), the First International Satellite Cloud Climatology Project (ISCCP) Regional Experiment-Arctic Cloud Experiment (FIRE-ACE) (Curry et al. 2000), the Mixed-Phase Arctic Cloud Experiment (M-PACE) (Verlinde et al. 2007), the Arctic Summer Cloud Ocean Study (ASCOS) (Tjernström et al. 2014), the Arctic Clouds in Summer Expedition (ACSE) (Tjernström et al. 2015), AFLUX and Surface Heat Budget of the Arctic Ocean (SHEBA) (Uttal et al. 2002). Taken together with data collected from ground-based remote sensing observations at Ny-Ålesund, cloud liquid and ice water contents appear to be

strongly influenced by synoptic conditions such as wind direction and the degree of thermodynamic coupling to the surface (Gierens et al. 2020).

The influence of aerosols on Arctic clouds was also documented based on observations from a large number of campaigns such as M-PACE (Prenni et al. 2009), the Arctic Study of Aerosol, Clouds and Radiation (ASTAR) (Yamagata et al. 2009), the Indirect and Semi-Direct Aerosol Campaign (ISDAC) (McFarquhar et al. 2011), the Aerosol-Cloud Coupling and Climate Interactions in the Arctic (ACCACIA) (Lloyd et al. 2015), the Aerosol, Radiation and Cloud Processes affecting Arctic Climate (ARCPAC) (Brock et al. 2011), PASCAL (Griesche et al. 2020), and the Polar Study using Aircraft, Remote Sensing, Surface Measurements and Models, of Climate, Chemistry, Aerosols, and Transport (POLARCAT) (Law et al. 2014), Radiation-Aerosol-Cloud Experiment in the Arctic (RACEPAC) (Herenz et al. 2018), the Vertical Distribution of Ice in Arctic Clouds (VERDI) (Klingebiel et al. 2015), and The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) (Jacob et al. 2010), which was one of the largest airborne field campaigns to study the impact of air pollution on Arctic climate. Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments (NETCARE) (Abbatt et al. 2019) was a highly interdisciplinary field campaign that was able to observe melt ponds as a source of dimethyl sulfide and long-range mineral dust as a prominent springtime source of INPs and local mineral dust as a local source in the summer. Arctic Mechanisms for the Interaction of the Surface and Atmosphere (AMISA) (Persson et al. 2017) was also a field campaign that complemented ASCOS with its information on the impact of synoptic and mesoscale flow and vertical mixing of aerosol particles. Additionally, International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) was a field campaign dedicated to determining the influence of local and transported aerosol particles on clouds among other effects such as haze and ozone over open ocean in the Arctic (Russell et al. 2010, Quinn et al. 2017, Huang & Jaegle 2017). Rare high-resolution airborne measurements of INPs and air temperature in the high Arctic were measured by the Polar Airborne Measurements and Arctic Regional Climate Model Simulation Project (PAMARCMiP) (Hartmann et al. 2019a) while evidence from airborne measurements during the Fifth Airborne Carbon Measurements (ACME-V) field campaign in the Alaska revealed that the Arctic is not always as pristine in the summer as once thought due to wildfires and local oil extraction activities (Creamean et al. 2018).

A number of field campaigns have also studied cloud-radiation impacts. Among the earliest to study Arctic surface energy fluxes were the Radiation and Eddy Flux Experiment (REFLEX) (Hartmann et al. 1991) and the Arctic Radiation and Turbulence Interaction Study (ARTIST) (Hartmann et al. 1999). Other studies with goals to better understand the impact of clouds on atmospheric radiation followed, including SHEBA (Stramler et al. 2011), ASCOS (Sedlar et al. 2011), the Solar Radiation and Phase Discrimination of Arctic Clouds (SoRPIC) (Bierwirth et al. 2013), the Arctic Radiation-IceBridge Sea & Ice Experiment (ARISE) (Smith et al. 2017) and ACLOUD/PASCAL (Stapf et al. 2021).

The influence of various surface types such as the Marginal Ice Zone (MIZ), melt ponds, leads and polynyas on cloud properties were also the interest of several Arctic field campaigns such as Measurements of Arctic Clouds, Snow, and Sea Ice nearby the Marginal Ice Zone (MACSSIMIZE), ACLOUD/PASCAL (Stapf et al. 2021), the recently completed

Multidisciplinary Drifting Observatory for the Study of Arctic Climate (MOSAIC) campaign, melt ponds on energy and momentum fluxes between atmosphere and ocean (MELTEX) (Roßel & Kaleschke 2012), Microbiology-Ocean-Cloud-Coupling in the High Arctic (MOCCHA), which aimed to quantify influences of new aerosol particle formation (Baccarini et al. 2020), particularly over leads, and PAMARCMiP. A summary of the impact of these various factors on clouds and the relevant field campaigns are summarized in Table 1.

3.2 Insights on Arctic cloud-controlling factors gained from ACLOUD, PASCAL and AFLUX

The influence of a number of cloud-controlling factors on Arctic low-clouds was investigated during ACLOUD, PASCAL (Wendisch et al. 2019) and ALFUX, which were components of Phase I of the German Arctic Amplification: Climate Relevant Atmospheric and Surface Processes and Feedback Mechanisms (AC³) field campaign (Wendisch et al. 2017). ACLOUD and PASCAL concurrently took place in May and June 2017 in and around Svalbard, while ALFUX took place between mid-March and mid-April 2019 in the same area. Here, we present a brief description of results from the campaigns to demonstrate the effectiveness of Arctic field campaigns in advancing our understanding of the factors driving cloud formation and cloud properties.

The influence of INPs on clouds and their dependence on the coupling state of the clouds to the surface was observed using ship-based remote sensing instruments taken during PASCAL. A high occurrence of surface-coupled ice-containing clouds with cloud-top temperatures warmer than -10°C suggests the influence of near-surface INPs on Arctic boundary layer clouds at relatively warm supercooled temperatures when the cloud layer is coupled to the surface.

The combination of ACLOUD/PASCAL and AFLUX, both of which took place in a MIZ revealed that the familiar cloud-free and cloudy atmospheric states observed during SHEBA over sea ice and snow-covered regions also occur over open ocean (Wendisch et al. 2019). The differences in the surface temperature and lapse rates between the ACLOUD/PASCAL and AFLUX field campaigns, which took place during different months, influence the clear and cloudy states. While the horizontal surface temperature gradient between sea ice and open ocean was 25 K in AFLUX, it was only 6 K in ACLOUD. The horizontal surface temperature gradients in turn affects the vertical lapse rate, i.e. thermodynamic stability of the atmosphere, which consequently affects downward LW emission profile in cloud-free conditions. Less stable atmospheric conditions decrease the net irradiances because less downward LW radiation is emitted from the atmosphere. Thus, the cloud-free modes over sea ice and open ocean may both shift in response to thermodynamic stability. Cloud-base temperature remains almost unchanged whether over sea ice or open ocean and, thus, downward LW radiation emitted by the cloud-base stays nearly constant. These shifts were revealed during the early-spring AFLUX (very stable) and the summer ACLOUD/PASCAL (less stable) campaigns.

ACLOUD and PASCAL also raised several open questions related to Arctic clouds and the factors that control them. Although clouds are clearly impacted by surface type, the degree to

which sea ice and open water impact cloud properties still remains poorly quantified. Moreover, observations revealed stronger turbulence between clouds at high altitudes than expected, raising the question of the dominant contributing physical processes leading to the enhanced turbulence. On the other hand, atmospheric thermodynamic stability was also observed to be weaker than previous studies suggest (Stapf et al. 2021).

4 Outlook

We have outlined and discussed the influence of various meteorological factors and aerosols, and the mechanisms by which they influence Arctic mixed-phase cloud properties. In so doing, we have identified several outstanding questions that remain to be addressed in the future to improve our understanding of the factors controlling the behaviour of Arctic mixed-phase clouds.

Progress in resolving these major questions in Arctic cloud evolution and their radiative effects is hindered by the limited number of high quality observations of cloud and aerosol processes. Satellite measurements have substantially advanced our current state of knowledge of Arctic cloud properties and their interaction with sea ice, however, both passive and active satellite observations suffer from a number of limitations. Current passive satellite data of cloud properties are limited by inaccurate retrievals at steep solar zenith angles that are exacerbated in the polar regions (Grosvenor & Wood 2014). While the development of new algorithms to correct for these biases related to the lack of three-dimensional radiative transfer effects has led to promising improvements (Lebsock & Su 2014, Khanal et al. 2020), there is still nontrivial disagreement in the various cloud properties among satellite instruments. The common supercooled liquid-topped structure of Arctic mixed-phase clouds also presents a challenge for active spaceborne lidar that cannot penetrate entire cloud layers with optical thicknesses greater than approximately 5 (Winker et al. 2009). Although the synergistic use of collocated measurements with spaceborne radar can remediate this shortcoming, the combination of instruments still fails to observe the bottommost kilometre of the atmosphere due to the combination of radar ground clutter and lidar beam attenuation (Liu et al. 2017). The horizontal and vertical spatial resolution of satellite observations is also insufficient to accurately determine the spatial distribution of clusters of liquid and ice structures that comprise mixed-phase clouds, which in turn impact the efficiency of the WBF process. Furthermore, spaceborne remote sensing instruments cannot reliably retrieve CCN and INP concentrations at a spatial resolution that is sufficient for cloud process modelling; this is particularly true for very small aerosol particles $< 0.1 \mu\text{m}$. To evaluate these issues dedicated validation exercises using ground and airborne measurements are required. While in situ observations are a more suitable tool for this purpose, they suffer from a lack of spatial coverage for the widespread low-level stratiform mixed-phase clouds that are ubiquitous in the Arctic (Eastman & Warren 2010). The lack of in situ observations is especially problematic during Arctic winter when harsh weather conditions prevail that can lead to aircraft icing during in situ measurements. In addition to clouds, tenuous aerosol layers are common in the Arctic and preclude measurements from spaceborne observations. Moreover, while the TOA radiative fluxes are better observed by satellite observations compared to surface fluxes (Kato et al. 2018), it is crucial to characterize surface

radiative energy fluxes for the important surface radiative energy budget and the related near-surface warming in the Arctic, which are difficult to retrieve from satellite data. Finally, although spaceborne infrared sounders have improved our knowledge of temperature and moisture inversions in the Arctic, their coarse vertical resolution precludes observations of shallow inversions. As a result of the limited high-quality observations, the precise mechanisms relating lower tropospheric stratification, cloud dynamics and vertical velocity are still poorly understood. We emphasize the need for reliable and comprehensive data of the response of Arctic mixed-phase clouds under a broad range of relevant meteorological and surface conditions. In this regard, the validation of models using data from dedicated measurement campaigns have a powerful potential to unravel model deficiencies in parameterizations, sub-scale process representation, and other issues. The validated models then reveal critical processes determining the evolution and effects of clouds.

Regarding the impact of aerosols on Arctic mixed-phase clouds, it is clear that Arctic meteorology and aerosol levels are continually undergoing dramatic changes. Local CCN and INP emissions will likely increase due to shipping and oil and gas development, mining, exposed soil from irreversible loss of snow, permafrost, and sea ice (Meredith et al. 2019), and altered sea spray and biogenic emissions from changes in sea ice cover, wind intensity and warmer temperatures (Arrigo 2008, Ardyna et al. 2014, Deslippe et al. 2012). A better understanding of continually changing natural aerosol emissions and the fundamental physical processes involved was emphasized to better constrain Arctic ACIs (Schmale et al. 2021). Aerosol transport from lower latitudes will also change with shifting wind and precipitation patterns, and there will likely be increasing sub-Arctic wildfire emissions and changing anthropogenic aerosol particle emissions as well. Drawing from the previous sections, we put forth ten recommendations to improve our understanding of cloud-controlling factors in the Arctic that would ideally involve the development of an overall community-wide strategic plan to improve on this front:

- Targeted field campaigns, dedicated model validations and model intercomparisons of synoptic influences such as cyclones, moisture intrusions and large-scale subsidence on clouds. In particular, cloud evolution and airmass transformations over Arctic sea ice and open ocean during moist intrusions, particularly from a Lagrangian perspective based on in situ observations are lacking yet important for model evaluation (Pithan et al. 2018, Neggers et al. 2019, Dimitrelos et al. 2020, Wendisch et al. 2021). This last point is the target of the upcoming HALO-(AC)³ field campaign planned to take place in 2022.
- Detailed investigations using high-resolution models to quantify the impact of surface aerosol and moisture sources versus cloud-top entrainment fluxes under various meteorological and surface conditions. High-resolution models are also needed to clarify the dominant planetary boundary layer processes affecting the in-cloud redistribution of CCN and INPs in mixed-phase clouds.
- Improved methods for observing INPs, CCN and ice particles in situ and to the extent possible, also from remote sensing measurements. The former task requires higher sensitivity to tenuous aerosol layers typical of the Arctic and accurate distinctions between INP and CCN types. Although limited accurate high-latitude (poleward of 82°N) aerosol particle measurements are available from passive satellite observations, they are currently unavailable from active spaceborne remote sensing instruments,

despite their increasing importance in a warming Arctic with decreasing sea ice extent. There has recently been active progress on in situ INP measurements. Year-long surface-based INP measurements at Oliktok that uses techniques described in (McCluskey et al. 2018) and (Suski et al. 2018) will soon be available and MOSAiC will provide the first year-round observations of Arctic INPs in the Arctic Ocean. However, these observations are limited to the surface and may not represent the cloud layer. The latter task requires improved retrieval algorithms for ice number concentration and ice crystal effective radius with reduced uncertainties in stratiform mixed-phase clouds. While such algorithms have been explored for ice number concentration using ground-based observations (Zhang et al. 2014), they are completely lacking using current satellite observations. This also includes further reduced shattering effects of ice crystals on aircraft measurements (Korolev et al. 2013).

- Improved understanding of ice formation and growth in Arctic mixed-phase clouds and representations of these processes in climate models. For example, representing subgrid-scale variability in the liquid and ice partitioning in mixed-phase clouds in climate models (Tan & Storelvmo 2016, Zhang et al. 2019) can result in more accurate rates of the WBF process in climate models and requires continuous and high spatial resolution observations of mixed-phase clouds. Detailed observations of snowflakes using three-dimensional ground-based cameras, e.g. the Multi-Angle Snowflake Camera (Garrett et al. 2012) in the Arctic are expected to aid in the development of more sophisticated parameterizations of ice cloud microphysical processes.
- Long-term observations that can be linked to multi-scale models, including in multi-layer cloud conditions. Furthermore, existing fair and consistent comparisons between models and satellite remote observations via the satellite simulator approach (Bodas-Salcedo et al. 2011) should not only be continued given their previous success in identifying model biases (Nam et al. 2012, Cesana et al. 2015), but also expanded to include other types of remote sensing instruments and a larger variety of observables. Ground-based satellite simulators (Kuma et al. 2021) are an example of a recent advance that has taken us one step closer to closing the gap between model and observation comparisons from the surface perspective, and the development of scale-aware and definition-aware diagnostics for near-surface precipitation frequency are also another example (Kay et al. 2018). The full potential of model and satellite comparisons is critical to reducing model biases but has yet to be fully exploited.
- Upgraded sophisticated methods to isolate the aerosol in observational studies from confounding factors such as co-varying meteorology and secondary ice formation; these methods are needed in the current and changing Arctic climate, including in response to sea ice decline.
- Boosted development and testing of Arctic aerosol transport models of dust and other aerosol particles, particularly over remote regions and in the presence of precipitation, along with better techniques for integrating satellite and suborbital data with models. These efforts could benefit from focused field campaigns that aim to validate Arctic aerosol transport models.

In summary, we have highlighted and reviewed a number of important Arctic cloud-controlling factors. The influences of these cloud-controlling factors share similarities yet are also markedly different from the impacts of tropical cloud-controlling factors (Klein et al. 2017). We contend

that a better understanding of the various controls over Arctic clouds is contingent on improved observations of clouds and aerosols in terms of both quality and quantity. Some of the shortcomings in the satellite instruments of the past and present are currently being considered in NASA's ATMosphere Observing System (ATMOS) mission resulting from the National Academies of Sciences, Engineering and Medicine's 2017 Decadal Survey (National Academies of Sciences and Medicine 2018). The launch of a spaceborne high spectral resolution lidar will enable higher sensitivity to tenuous aerosol layers. Additionally, while currently still under development, coincident observations of aerosols and clouds in the Arctic by exploiting far-infrared measurements as well as improved observations of cloud ice microphysics and snowfall are being considered. Due to the previous success of active satellite instruments in improving our understanding of cloud processes and better constraining cloud feedbacks (Winker et al. 2017), particularly in the Arctic (Kay & Gettelman 2009, Taylor et al. 2015, Morrison et al. 2018), active satellite observations are being considered in the ACCP mission. Combining these observations with targeted field campaigns presents a path forward to closing the gap in our knowledge of the controls over Arctic clouds and therefore enable a better understanding of the role of clouds in the changing Arctic climate system.

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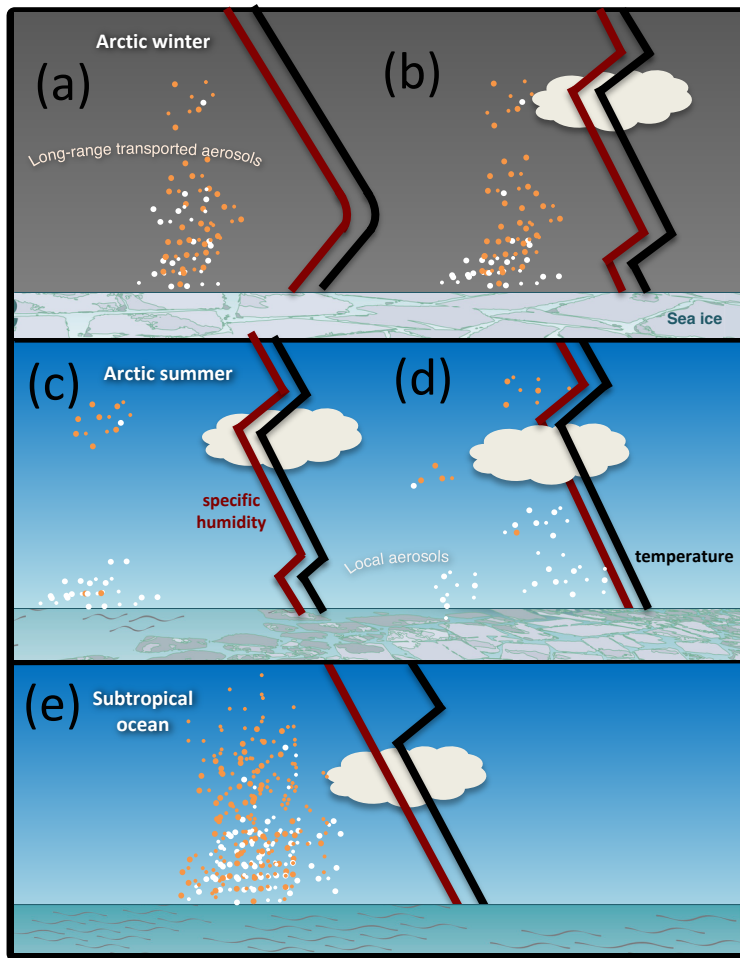
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1918

1919 **Figure 1.** Schematic of typical thermodynamic structures of the Arctic atmosphere during (a)
 1920 winter in the presence of clear-sky or thin ice clouds, (b) winter in the presence of mixed-phase
 1921 clouds, (c) summer when clouds are decoupled from the surface, (d) summer when clouds are
 1922 coupled to the surface (e) subtropical stratocumulus clouds. The dashed (solid) lines indicate
 1923 specific humidity (temperature) profiles and the triangles indicate local aerosol particles that are
 1924 overall less abundant than long-range transported aerosol particles (dots) in the winter. Coupling
 1925 of the clouds to the surface facilitates interactions with more local aerosol particles. Overall,

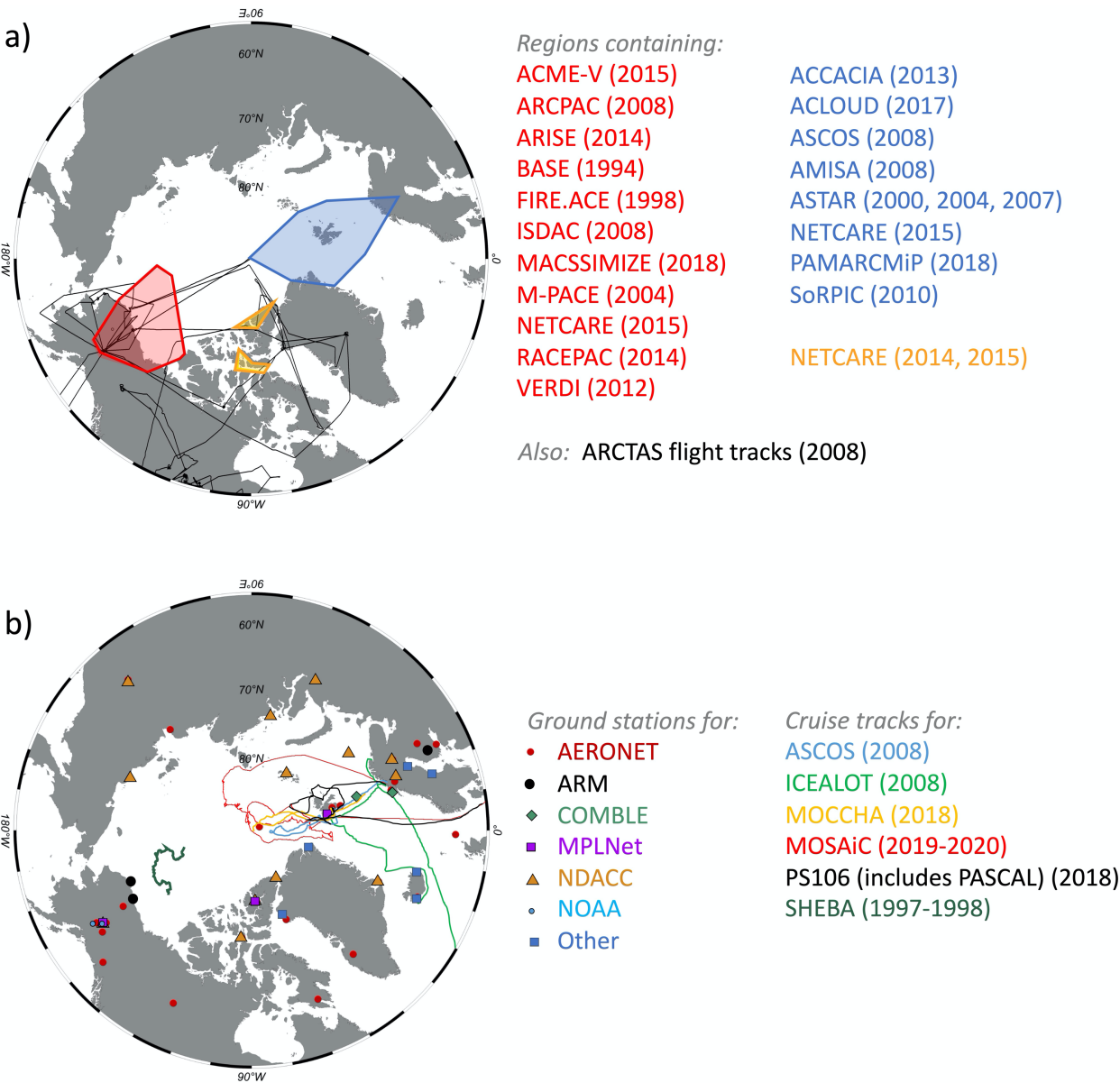


Figure 2. Locations of selected Arctic campaigns and datasets, including **a)** aircraft datasets, and **b)** ground and ship-based datasets.

1936 **Table 1.** List of Arctic field campaigns and the cloud0controlling factors that were observed
 1937

Meteorological variable	Surface type	Relevant field campaign
Thermodynamic structure		BASE, FIRE-ACE, M-PACE, ASCOS, ACSE, AFLUX, SHEBA
Moisture intrusions		SHEBA, ACCACIA, ACLOUD, PASCAL, MOSAiC
Aerosol particles		M-pace, AMISA, ASTAR, ISDAC, ACCACIA, ARCTAS, ARCPAC, POLARCAT, VERDI, ICEALOT, RACEPAC, ACME-V, NETCARE
	Marginal Ice Zone (MIZ)	ACSE, ACLOUD, PASCAL,
	Melt ponds	MACCSIMIZE,
	Polynya	NETCARE
	Leads	MELTEX, NETCARE
		PAMARCMiP
		ASCOS, MOCCHA,
		PAMARCMiP

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