Chapter 1. Introduction

The Arctic region has experienced *disproportionate* climate warming in recent decades, a phenomenon known as Arctic amplification. Observations show that surface temperatures in the Arctic are rising at about three to four times the global average rate (Rantanen et al., 2022; Vihma, 2014). This accelerated warming has led to dramatic losses in sea ice extent and thickness. Late-summer Arctic Sea ice coverage has declined by roughly 13% per decade since 1979 (Serreze & Stroeve, 2018), with record minima in recent years. Sea ice loss triggers well-known feedback loops: as highly reflective ice is replaced by dark open water, more solar energy is absorbed, further warming the ocean and atmosphere (the ice–albedo feedback). Reduced ice cover also influences vertical heat fluxes and moisture supply to the atmosphere. These changes can modify atmospheric circulation and precipitation patterns at high latitudes, with potential impacts extending to mid-latitude weather extremes (Vihma, 2014; Screen et al., 2018). The interplay of these processes makes the Arctic a focal point for studying climate feedbacks and uncertainties in future climate projections.

Among the lesser-understood aspects of Arctic amplification are the roles of clouds and aerosols in modulating the surface energy budget. Arctic clouds are unique in their properties and radiative effects. In winter, when polar nights prevail, clouds act

primarily to insulate the surface by trapping outgoing longwave radiation, producing a warming influence (Shupe & Intrieri, 2004). In summer, the nearly continuous daylight means that low clouds increase the planetary albedo and exert a net cooling effect by reflecting solar radiation. The net impact of clouds on Arctic surface temperature thus depends on cloud properties, season, and surface conditions. As the climate warms and sea ice retreats, changes in cloud cover and cloud phase (liquid vs. ice) could either amplify or dampen Arctic warming. For instance, an increase in low-level liquid-containing clouds over newly open water could reflect more sunlight in spring and summer, offsetting some warming, whereas in autumn or winter such clouds might enhance downwelling longwave flux, promoting warming. Recent satellite-based analysis by Cesana et al. (2024) provides evidence of a *negative cloud phase feedback* associated with sea ice loss: as sea ice cover decreases, the frequency of low-level liquid-bearing clouds increases, and these clouds were found to cool the surface by reflecting more shortwave radiation, with negligible longwave warming effect. Cesana et al. (2024) showed that in the present climate this mechanism is strongest in the colder seasons and could act to dampen future Arctic surface warming as sea ice continues to decline. This intriguing result underscores that accurate representation of cloud phase and cloud–radiation interactions is essential for reliable Arctic climate projections.

At the heart of cloud formation and radiative effects are atmospheric aerosols – tiny particles that act as seeds for cloud droplets and ice crystals (cloud condensation nuclei, CCN, and ice-nucleating particles). The Arctic atmosphere has historically been very clean in terms of aerosol burden, especially in summer, due to its remoteness from human pollution sources. However, aerosol concentrations show strong seasonal variability. In winter and early spring, the Arctic experiences "Arctic haze," a period of enhanced aerosol loading attributed to transport of mid-latitude anthropogenic pollution under stable atmospheric conditions. In summer, by contrast, aerosol number concentrations drop to very low levels (often <100 cm^{-3} accumulation-mode particles), and local natural aerosol sources become relatively more important (Willis et al., 2018; Schmale et al., 2021). These natural sources include marine aerosols such as sea salt from open water (sea spray) and sulfate particles from biogenic gases. The extraordinarily pristine summer atmosphere means cloud microphysics can be highly sensitive to slight changes in aerosol availability. A longstanding hypothesis in climate science, the CLAW hypothesis (Charlson et al., 1987), posits that marine ecosystems could regulate climate via emissions of dimethyl sulfide (DMS), a biogenic sulfur gas. DMS oxidizes in the atmosphere to form sulfate aerosol, which can act as CCN and potentially increase cloud reflectivity (thus cooling the surface). In the Arctic context, as sea ice retreats and marine biological activity increases, DMS emissions may rise, leading to more

cloud-forming aerosols and altering cloud properties (Shaw, 1983; Ghahreman et al., 2019). Parallel feedback might occur with sea spray aerosols: more open water in a warmer Arctic allows greater production of sea salt particles by wind and wave action. Sea spray can directly scatter sunlight and also serve as CCN. Additionally, new aerosol particle formation from marine precursors in the Arctic may contribute numerous ultrafine particles that grow into the CCN size range under the clean, supersaturated conditions prevalent in Arctic low clouds (Leaitch et al., 2016; Karlsson et al., 2021). The net effect of these aerosol-cloud interactions on Arctic amplification could be significant yet is poorly constrained. Will increased natural aerosol emissions counteract warming by brightening clouds (a negative feedback), or could certain aerosol influences actually enhance warming (for example, if clouds increase in winter or if absorbing aerosol settles on snow/ice)? These questions motivate careful investigation of aerosol–cloud–radiation interactions in the rapidly changing Arctic.

Current global Earth system models (ESMs) – including those contributing to the Coupled Model Intercomparison Project Phase 6 (CMIP6) – face challenges in capturing the complex Arctic aerosol and cloud processes. Many CMIP6 models struggle to reproduce observed Arctic cloud amounts, aerosol concentrations, and the timing of seasonal changes (Lauer et al., 2022; Webster et al., 2022). Notably, a recent evaluation of marine aerosol in 12 CMIP6 models by Lapere et al. (2023)

revealed substantial biases in polar regions. They found that models overestimate sea salt aerosol mass concentrations at the surface by roughly an order of magnitude compared to observations in both the Arctic and Antarctic. The models also exhibit large inter-model spread (uncertainty up to a factor of ~3) in polar aerosol levels (Lapere et al., 2023). These discrepancies point to missing or misrepresented processes – for example, *localized aerosol sources tied to sea ice*. Many models do not explicitly account for aerosols emitted from leads (fractures in sea ice) or from the sublimation of salty blowing snow, leading to distorted aerosol seasonal cycles at high latitudes (Lapere et al., 2023; Huang et al., 2018). Indeed, Lapere et al. (2023) suggest that the absence of these polar-specific aerosol sources causes modelled aerosol concentrations to peak at the wrong time of year and fail to match the observed summertime increases near the poles. Such model shortcomings directly translate into uncertainties in cloud simulation and surface radiation. The net effect can be a sizeable error in the Arctic surface energy budget – on the order of 1 W m^{-2} in present-day climate, according to Lapere et al. (2023). For context, 1 W m^{-2} is comparable to the perturbation by some anthropogenic forcings, meaning that poorly represented natural aerosols could noticeably skew Arctic climate projections.

Another critical aspect is how models handle future changes in these aerosols. With continued sea ice decline, natural marine aerosol emissions are expected to increase. Marine biologically-derived sulfate (from DMS) and primary sea spray could double

in concentration by 2100 in the Arctic under high-emission scenarios, and even under moderate climate change scenarios at least a ~50% increase is projected (Lapere et al., 2023). This implies that the associated cloud radiative effects – either cooling or warming – could become more pronounced in the future Arctic. Yet, if models do not capture the baseline processes correctly, their future predictions of Arctic amplification may be off the mark. Additionally, recent analyses of CMIP5 vs CMIP6 indicate that the latest generation of models shows *higher equilibrium climate sensitivities* on average, largely due to stronger positive cloud feedbacks (Zelinka et al., 2020). Differences in how models treat cloud phase and microphysics, especially in stratiform clouds, contribute to a wide range in projected Arctic cloud changes (Bock & Lauer, 2024). High-ECS models in CMIP6 tend to simulate smaller increases (or even decreases) in the cooling effect of polar clouds under warming, whereas lower-ECS models simulate clouds that produce larger negative feedback (Bock & Lauer, 2024). This divergence hints that some models might be missing negative feedback processes such as those involving aerosol-cloud interactions that stabilize the climate. In summary, there is growing recognition that improving the representation of natural aerosol–cloud processes in the Arctic is key to narrowing uncertainties in climate projections (Thornhill et al., 2021; Willis et al., 2023).

Scientific Motivation and Objectives: Given this context, the motivation for the present work is to investigate how natural marine aerosols – in particular dimethyl sulfide (DMS)-derived sulfate aerosols and sea spray aerosols – influence Arctic cloud radiative forcing and feedbacks, and how well these processes are captured in state-of-the-art models. The overarching hypothesis is that enhanced emissions of marine aerosols from a warming, ice-free Arctic Ocean could provide a negative climate feedback by increasing cloud reflectivity (especially in summer), partially mitigating Arctic amplification. However, verifying this hypothesis requires answering several specific research questions:

- *RQ1:* How do marine biogenic aerosol sources, such as DMS emissions, contribute to cloud condensation nuclei populations and cloud properties in the Arctic? For example, what fraction of Arctic cloud droplet nuclei can be traced to DMS-derived sulfate, and does this lead to significant changes in cloud brightness (albedo)?
- *RQ2:* What is the impact of increased open-water sea spray aerosol on low-level Arctic clouds and the surface energy budget? Does sea spray from newly opened waters or leads enhance cloud cover or liquid water content, and if so, in which seasons is this effect most important?
- *RQ3:* Do current Earth system models (CMIP6) adequately simulate these aerosol-cloud interactions? This involves assessing biases in aerosol

concentrations (sulfate, sea salt) and cloud radiative effects in models, and determining if model experiments that include or exclude DMS/sea-salt emissions show differences in Arctic cloud radiative forcing.

- *RQ4:* Could improve representation of natural aerosol processes reduce uncertainty in Arctic climate projections? In other words, if models incorporated refined aerosol emissions schemes (e.g., accounting for leads, updated DMS fluxes), would the spread in projected Arctic warming and sea ice loss decrease or the mean projection change?

To address these questions, this study will leverage data and simulations from the AerChemMIP subset of CMIP6, which includes specialized experiments isolating aerosol and chemistry effects. By analyzing model runs with and without natural aerosol sources (for instance, pre-industrial vs present-day DMS emissions, or sensitivity tests removing marine sources), we can infer the radiative forcing due to these aerosols and evaluate their role in Arctic climate feedbacks. Observational datasets (satellite retrievals, surface measurements from Arctic field campaigns) will be used as a benchmark to evaluate model skill in reproducing aerosol and cloud properties.

The significance of this project lies in improving our understanding of a potentially important *negative feedback mechanism* in the Arctic. If marine aerosol-cloud

interactions are found to dampen warming, they represent a natural brake on Arctic amplification that is currently under-represented in models. Conversely, if these processes are negligible or counteracted by other factors, that too is vital to ascertain. In either case, the findings can guide model development: for instance, informing parameterizations for sea-ice-related aerosol emissions or cloud microphysics in polar conditions. Reducing model biases in the Arctic is crucial, as this region is a bellwether for global climate change and a key determinant of sea-level rise (through Greenland melt) and mid-latitude weather linkages.

Outline of the Thesis: This thesis is organized into six chapters. Following this introduction, Chapter 2 provides a review of the relevant literature and current state of knowledge on Arctic amplification, aerosol-cloud-radiation processes, and model performance, highlighting gaps that motivate our study. Chapter 3 describes the data and methodology, including the climate model experiments analyzed (CMIP6 AerChemMIP simulations) and the observational datasets and metrics used for evaluation. Chapter 4 presents the results of our analysis, focusing on the influence of DMS and sea spray aerosols on cloud properties and radiative fluxes in the Arctic, as well as how models compare to observations. Chapter 5 discusses the implications of these results, examining uncertainties, potential feedbacks, and how our findings fit into the broader context of Arctic climate dynamics. Finally, Chapter 6 concludes

the thesis by summarizing the key findings and contributions, and offering recommendations for future research and model improvements in this domain.

Chapter 2. State of the Art and Bibliography

2.1 Arctic Amplification and the Role of Sea Ice and Clouds

Arctic amplification – the enhanced warming of the Arctic relative to the global average – is a well-established phenomenon with multiple contributing mechanisms. The most prominent driver is the ice–albedo feedback: as rising temperatures melt snow and sea ice, darker ocean and land surfaces are exposed, leading to greater absorption of solar radiation and further warming. This basic feedback has been quantified in numerous studies and is estimated to account for a substantial fraction of the Arctic's excess warming (Pithan & Mauritsen, 2014). However, feedbacks involving atmospheric temperature, water vapor, and clouds also play critical roles. The Arctic tends to have a strong lapse rate feedback – as the surface warms, the vertical temperature profile adjusts in a way that enhances radiation to space less efficiently than in the tropics, thus trapping more heat (Goosse et al., 2018). Additionally, increased moisture in the Arctic air (from local evaporation and poleward transport) can amplify warming through the water vapor feedback and by increasing cloud cover (Bintanja & Selten, 2014).

Clouds in particular represent a complex feedback component. Because the Arctic surface, especially when ice-covered, is very cold and often close to the temperature of low clouds, the net cloud radiative effect can swing between warming and cooling

depending on season and cloud characteristics. In winter, clouds generally warm the surface by providing a downwelling longwave flux that exceeds the negligible solar flux, whereas in summer, clouds have a net cooling effect by reflecting solar radiation (intriguingly, this cooling effect can locally counteract the ice–albedo feedback to some extent). The balance between these effects is sensitive to cloud microphysics – notably the phase (liquid or ice) and particle concentrations. Mixed-phase clouds (containing supercooled liquid droplets) are prevalent in the Arctic and have high emissivity and albedo. An increase in liquid-containing clouds in spring/summer would raise the albedo (strong cooling impact), while the same increase in autumn/winter would raise the greenhouse effect (warming impact).

Recent satellite analyses and model studies have started to quantify these cloud-phase feedbacks. Cesana et al. (2024) used A-Train satellite observations to correlate sea ice concentration (SIC) with cloud properties and surface radiation. They found a robust inverse relationship between SIC and low-level liquid-bearing cloud cover: as sea ice decreases, the occurrence of low clouds with liquid water increases (especially in the lowest ~3 km). This was evident in all seasons except the peak of summer, with the strongest signal in winter. Importantly, Cesana et al. noted that the *radiative effect* of these extra clouds was an enhanced cooling at the surface – a perhaps counter-intuitive result for wintertime. The explanation is that the newly formed low clouds reflect much more solar radiation (when sun is above the horizon)

than the dark open water beneath would, thus cooling the surface, and their longwave emission downward is roughly on par with the longwave emission that was coming from the warmer open water surface (meaning the clouds did not add much extra greenhouse effect). In essence, the net effect of replacing sea ice with open water and low clouds was surface cooling in their observational analysis, implying negative feedback on warming (Cesana et al., 2024). This finding aligns with earlier model suggestions that polar cloud adjustments could dampen sea-ice-driven warming (e.g., Alkama et al., 2020, who also suggested summer cloud increases might offset some sea ice albedo effect). It should be noted that these effects are sensitive to cloud optical properties and the seasonal cycle of insolation – thus, models must handle cloud microphysics accurately to capture this feedback.

Despite these nuances, many climate models historically struggled with Arctic clouds. They often either underpredict cloud fraction or misrepresent the phase partitioning (e.g., having too much ice and too little supercooled liquid, which affects radiative impact). Earlier model intercomparisons (e.g., CMIP5) revealed large spread in Arctic cloud feedbacks, contributing to uncertainty in Arctic amplification projections (Vavrus et al., 2016). In the current generation, CMIP6, some models exhibit *very high equilibrium climate sensitivity (ECS)* values in part due to strong positive cloud feedbacks at low latitudes (Zelinka et al., 2020). However, differences in Arctic cloud response are also evident. Bock and Lauer (2024) analyzed 51

CMIP5/CMIP6 models grouped by low, medium, and high ECS. They found that high-ECS models show a smaller increase in the cooling effect of polar clouds under warming compared to low-ECS models. In practical terms, high-ECS models did not boost Arctic cloud reflectivity as much as low-ECS models did in a warmer climate, implying that high-ECS models might be missing a negative cloud phase feedback (or conversely, low-ECS models might be exaggerating it). This result underscores that *modeling of Arctic cloud-phase feedbacks remains an open research area*. It is exactly in this context – where clouds and sea ice interact – that aerosols enter the picture, since aerosol availability can determine cloud microphysical properties (droplet number, size, phase propensity) and thereby cloud radiative behavior.

2.2 Arctic Aerosol Sources: Anthropogenic Haze vs. Natural Marine Aerosols

The Arctic aerosol life cycle is markedly different from that at lower latitudes, featuring extreme seasonal swings and a mix of distant and local sources. In winter and early spring, the phenomenon of Arctic haze occurs: the lower Arctic troposphere accumulates aerosol and pollution transported from mid-latitude industrial regions of Europe, Asia, and North America. These particles (sulfate, black carbon, etc.) can remain trapped under temperature inversion layers for weeks, leading to a persistent haze (Shaw, 1983; Quinn et al., 2007). Arctic haze has been studied since the 1980s and is known to increase aerosol optical depth and have a

minor warming effect (due to absorption by soot and trapping of outgoing longwave), although its overall climate impact is modest compared to greenhouse gases. Anthropogenic aerosol emissions have declined in recent decades (especially sulfur from Europe/N. America), and accordingly Arctic sulfate aerosol in spring has shown some decreasing trends (Law et al., 2017). Even so, the haze period remains the time of *maximum aerosol loading* in the Arctic year.

In contrast, the summer Arctic atmosphere is exceptionally pristine. During summer, efficient wet removal (frequent cloud and fog formation leading to precipitation) and the general blocking of long-range transport by changes in circulation leave the Arctic with very low aerosol concentrations. Number concentrations of CCN-sized particles (>~100 nm diameter) can drop below 10 –50 cm^{-3} in the central Arctic boundary layer (Heintzenberg et al., 2015). Under such clean conditions, cloud microphysics can become aerosol-limited – meaning that an increase or decrease in available CCN can significantly alter cloud droplet number and size, and hence cloud optical thickness and longevity (Mauritsen et al., 2011). This is a regime quite unlike polluted regions where clouds have an abundance of CCN. Field observations confirm that the summer Arctic aerosol population is dominated by ultrafine and Aitken mode particles (diameters < 100 nm), many of which are believed to originate from secondary formation (new particle formation from precursor gases followed by growth) rather than direct emission (Heintzenberg et al., 2015; Croft et al., 2016).

The main precursor gases for particle formation in the Arctic include sulfur species (notably dimethyl sulfide, DMS, oxidized to sulfuric acid) and volatile organics from marine or terrestrial biospheres. Additionally, trace gases like iodine (released from surface seawater or ice algae) can nucleate particles; iodine oxide has been implicated in new particle formation during the spring–summer transition (Baccarini et al., 2018).

Marine biogenic aerosols are particularly important in summer. DMS, produced by phytoplankton in open water and in the marginal ice zone, is the dominant natural sulfur source. It has long been considered a climate-regulating agent (Charlson et al., 1987). Recent work by Ghahreman et al. (2019) demonstrated that DMS is indeed the main source of biogenic sulfate aerosol in the Arctic atmosphere during summer. They implemented DMS chemistry in a regional atmospheric model for the Canadian Arctic and found that including DMS increased summertime sulfate aerosol mass by up to 100% in parts of the Arctic, with most of the increase occurring in particles 50–200 nm in diameter – sizes that can act as CCN. This result emphasizes that DMS emissions can substantially boost the aerosol that seeds cloud droplets. Moreover, the production of DMS is tied to open water area and biological productivity; as sea ice recedes and the ocean warms, DMS emissions are expected to rise (Galí et al., 2019). Indeed, observations indicate a trend of increasing DMS flux in some Arctic regions over the past decades correlating with reduced ice cover

(Jarníková et al., 2018). However, accurately simulating DMS remains challenging because it involves coupling ocean biogeochemistry with atmospheric chemistry. There are uncertainties in how quickly marine ecosystems will respond (e.g., will new species with different DMS output dominate?) and how clouds respond to the extra sulfate.

Another key natural source is sea spray aerosol (SSA), composed mainly of sea salt (sodium chloride) and organic matter from the ocean surface. In the open ocean, SSA is produced by breaking waves (whitecaps) and bubble bursting processes, with emission flux strongly dependent on wind speed at the sea surface. The Arctic Ocean, when covered by ice, has limited areas for such production, but as the ice cover diminishes in summer, the open water fraction increases and so does potential SSA emission. Additionally, within the pack ice, leads (elongated cracks or openings in sea ice) can be significant localized sources of SSA even during winter. Measurements have shown that leads, despite their small fractional area, can generate aerosols with characteristics distinct from open-ocean spray – often enriched in organics and with different size distributions (Nilsson et al., 2001; Leck et al., 2002). The presence of young ice and brine on leads may allow bubble bursting at lower wind speeds or via thermal/bubble mechanisms unrelated to whitecaps (Nilsson et al., 2001). Ioannidis et al. (2023) recently modeled wintertime sea-spray aerosols under Arctic haze conditions and found evidence that open leads were the

primary source of fresh sea-salt particles in the coastal Arctic (near Utqiaġvik, Alaska) during winter, far exceeding contributions from long-range transported sea salt. Their WRF-Chem simulations indicated that standard open-ocean SSA source functions underestimate Arctic winter aerosol unless leads and blowing snow sources are included. The model, after updating the SSA emissions to account for lower temperatures and some organics, showed improved agreement with observed aerosol number concentrations (Ioannidis et al., 2023). This points to the crucial role of even small open-water fractions in seeding the Arctic atmosphere with particles.

Blowing snow-derived aerosol is another exotic Arctic source: strong winds can loft saline snow particles from sea ice, and as these sublimination they release sea salt aerosol. This process can dominate sea-salt aerosol production during cold, dark months over sea ice (Yang et al., 2019). While our focus is marine (open water) aerosols, it is worth noting that blowing snow is a natural source closely tied to sea-ice conditions and is not typically included in standard global aerosol models either.

In summary, the natural aerosol baseline in the Arctic is controlled by marine-related processes – biogenic sulfur emissions, open water and lead-derived sea spray, and to some extent blowing salty snow. These sources are highly sensitive to climate change: more open water (both spatially and temporally) generally means more aerosol precursor emissions. At the same time, a changing Arctic might introduce

new factors (e.g., increased terrestrial aerosol from thawing permafrost regions or wildfire smoke transport in summer). But broadly, as anthropogenic Arctic haze is expected to continue diminishing (with pollution controls) and the Arctic Ocean opens up, the aerosol regime is likely to become *more dominated by natural aerosols*. This transition is already being observed as a shift in Arctic aerosol seasonality: summer and autumn aerosol levels are not as low as they once were, and the composition is showing higher contributions from marine sources (Willis et al., 2019; Abbott et al., 2022). Willis et al. (2023) provides a comprehensive review of polar ocean–atmosphere interactions and note that while we have improved understanding of some processes, *the environmental drivers of natural Arctic aerosol variability remain poorly quantified*. In particular, Willis et al. highlight those processes leading to aerosol emissions (such as biological activity bursts, ocean surface microlayer dynamics, etc.) are not well represented in models (Willis et al., 2023; Schmale et al., 2021). This uncertainty in natural aerosol drivers translates to uncertainty in how clouds and climate will respond in the Arctic.

2.3 Aerosol–Cloud–Radiation Interactions in the Arctic

Aerosols influence cloud properties in two main ways: the microphysical pathway (acting as CCN or ice nuclei, thereby changing cloud droplet number and size) and the radiative/thermodynamic pathway (absorbing or scattering radiation, or altering

atmospheric stability). In the Arctic, the microphysical effects are particularly significant given the low background CCN concentrations. An increase in CCN from natural aerosol sources can lead to clouds with more numerous but smaller droplets (assuming liquid water content stays similar). According to classical aerosol indirect effect theory (Twomey, 1977), such clouds are more reflective (higher albedo) – the so-called first indirect effect. Additionally, smaller droplets suppress drizzle and prolong cloud lifetime (the second indirect or Albrecht effect), which can maintain cloud cover longer (Albrecht, 1989). In the Arctic summertime, these effects could enhance the cooling influence of clouds by keeping clouds optically thick and persistent, thus reflecting sunlight over longer periods.

There is observational evidence consistent with these ideas. The low marine aerosol conditions sometimes coincided with very low cloud droplet numbers, potentially allowing more solar radiation through (Lance et al., 2011). Conversely, slight increases in aerosol (from either a pollution plume or a burst of marine particles) would noticeably increase cloud droplet concentration. Mauritsen et al. (2011) famously documented a case of a thin stratus cloud persisting for days over the central Arctic Ocean – a so-called "self-maintaining" cloud – and hypothesized that low turbulence and low CCN supply allowed it to exist in a delicate balance (any precipitation that fell scavenged enough droplets to reduce rain and thus prolong the

cloud). This underscores that aerosol-cloud microphysics in the Arctic can operate in a regime different from typical midlatitude clouds.

From the modeling perspective, Price et al. (2023) (as cited in Elementa by Pitzel et al., 2023) conducted an idealized modeling study to examine how the presence of aerosol just above the Arctic boundary layer can affect low-level cloud sustenance. They based their scenario on observations of enhanced aerosol in the lower free troposphere (from tethered balloon profiles; Lonardi et al., 2022) and investigated a situation with extremely low surface CCN (~1 cm^{-3}) but higher aerosol aloft. The simulations showed that when the concentration of above-cloud CCN was increased (up to 50× background), Arctic stratocumulus clouds could persist much longer. The mechanism is that entrainment of aerosol from the free troposphere into the cloud layer compensates for the lack of surface CCN, maintaining droplet numbers and preventing the cloud from dissipating due to gravitational settling and precipitation. Essentially, entrained aerosols can serve as a continuous source of CCN for the cloud. This result by Price et al. (2023) highlights an important pathway: even if surface-level aerosol is near zero (as often observed in summer), a reservoir of particles above inversion, perhaps from long-range transport or upper-level new particle formation, can mix down and keep clouds "alive." Igel et al. (2017) had earlier pointed out a similar idea that Aitken mode particles (very small ~30 nm) might get activated in supersaturated Arctic clouds and thereby contribute to

sustaining clouds when larger accumulation-mode particles are scarce. The Price et al. study gives modeling credence to that concept. It also suggests that models need to represent vertical aerosol distribution and entrainment accurately; if a model immediately scavenges all aerosol or lacks vertical transport, it might artificially kill clouds too quickly in clean environments.

Another facet of aerosol-cloud interaction is the glaciation of clouds. If aerosols acting as ice-nucleating particles (INPs) increase, they can trigger earlier ice formation in supercooled clouds, potentially causing the cloud to precipitate (snow out) and diminish – a warming impact if it reduces cloud cover (since less solar reflection). The Arctic is known for mixed-phase clouds that exist in a delicate balance of liquid and ice. Aerosols of different types (mineral dust, biological particles) can act as INPs. There is ongoing research into how aerosol changes might alter cloud phase partitioning. For example, Tan et al. (2022) found that an increased influx of Asian dust in late spring can enhance ice in Arctic clouds, potentially reducing cloud optical thickness. Though not the focus of our marine aerosol angle, it's worth noting as part of the state of knowledge that aerosol impacts on cloud phase are another source of uncertainty.

When considering direct radiative effects, Arctic aerosols generally exert a cooling by scattering sunlight (sulfate, sea salt) except for black carbon which can cause

local warming (especially when deposited on snow/ice, darkening it). However, the direct radiative forcing of natural aerosols like sulfate and sea salt in the Arctic is relatively small in magnitude compared to their indirect effects, primarily because solar radiation is limited much of the year and these aerosols are not strongly absorbing. Thornhill et al. (2021) in a multi-model study estimated the total aerosol effective radiative forcing (ERF) from preindustrial to present (global) as around –1.0 W m^{-2} (cooling), with a range across models. The Arctic portion of that forcing is a small fraction, but what's interesting is the potential *future* aerosol forcing. As anthropogenic aerosols continue to decline (a positive forcing, i.e., unmasking greenhouse warming), natural aerosol changes could either offset or add to that. For instance, if marine aerosol emissions increase in the future Arctic, they would produce a negative forcing (cooling influence) regionally. One study by Acosta et al. (2021) using a global model projected that the cloud radiative effect of DMS-driven aerosol in the Arctic could grow more negative (stronger cooling) by end-of-century, partially counteracting the warming from greenhouse gases in that region – though not enough to halt amplification, it could trim a few percent off the temperature rise.

In summary, aerosol-cloud-radiation interactions in the Arctic are characterized by an outsized influence of small changes in aerosol on cloud radiative properties. The sign of the resulting climate feedback is likely negative in summer (more aerosol →

more clouds → cooling) and could be neutral or slightly positive in winter (more aerosol → perhaps more clouds → warming). The annual net effect is uncertain. What is clear from the literature is that insufficient understanding of these processes remains a major knowledge gap. As noted by Willis et al. (2018) in a review of Arctic aerosol, processes controlling Arctic aerosol composition and abundance involve numerous components (ocean biology, sea ice physics, atmospheric chemistry, transport) and hence are difficult to capture in models. Schmale et al. (2021) likewise emphasize that Arctic aerosol processes are changing and that our predictive capability for aerosol-cloud interactions in the Arctic is limited by lack of observations and incomplete process representation in models.

2.4 Representation in Climate Models: CMIP6 Advances and Limitations

Contemporary Earth system models (ESMs) have made strides in simulating Arctic climate, but significant deficiencies remain in the context of aerosols and clouds. The CMIP6 suite introduced higher spatial resolutions and updated physics, and some systematic improvements over CMIP5 were documented. For example, Lauer et al. (2022) report that the multi-model mean total cloud cover and cloud water path in CMIP6 shows slightly smaller biases compared to CMIP5 when validated against satellite data, and pattern correlations of cloud fields improved (i.e., CMIP6 models on average got closer to observed cloud climatology). They attribute this to updated

cloud microphysics and tuning that occurred between CMIP5 and CMIP6. In particular, the notorious bias of excessive low-level cloud over the Southern Ocean was reduced in CMIP6, hinting at progress in cloud parameterizations.

However, when it comes to the Arctic, many CMIP6 models still do not capture critical features. Lauer et al. (2022) found that even in CMIP6, there are persistent biases in cloud radiative effect at high latitudes. Total cloud fraction in the Arctic is often underestimated in models, and the partitioning between liquid and ice in mixed-phase clouds is frequently biased towards too much ice, leading to clouds that are optically thinner than observed (Cesana et al., 2019; Tan et al., 2019). These microphysics issues cause models to misjudge the cloud feedbacks discussed earlier.

A particularly glaring model deficiency, as introduced in Chapter 1, is the treatment of natural aerosol sources in polar regions. Until recently, global models have typically not included sea-ice-related aerosol emissions (such as blowing snow salt or lead-specific fluxes). They use open-ocean empirical whitecap formulas (e.g., Monahan's or Gong's function) scaled by the fraction of open water in a grid cell. This approach might severely under-represent emissions from small leads or from newly ice-free areas with unique oceanographic conditions (Lapere et al., 2024). Lapere et al. (2023) specifically evaluated the CMIP6 models' performance for sea salt aerosol in polar regions. They discovered a paradox: despite missing polar

sources, CMIP6 models showed higher sea salt aerosol mass near the surface than observed (a factor of 10 high, as mentioned). The reason is that these models were calibrated to match global aerosol optical depth and had too high emissions or too slow removal of sea salt generally. But the seasonal cycle was wrong – models often showed peak sea salt in winter when winds are strong, whereas observations at Arctic sites often show peaks in late spring or summer, suggesting contributions from seasonal processes like biogenic organics or more open water later in the season. Lapere et al. (2023) conjectured that model "fill in" the missing summer source by overestimating background sea salt. They demonstrated with proxy calculations that adding sea-ice-related sources (like blowing snow or lead emissions) could better reproduce the observed spring/summer sea salt peaks and reduce the overestimation in winter. In effect, models might need to *redistribute* aerosol emissions in time and space – less "open ocean" sea salt in winter (when the ocean is actually frozen) and more local sea ice source aerosol in spring/summer.

Another relevant CMIP6 initiative is the AerChemMIP experiments, which aim to isolate the effects of aerosols and reactive gases. AerChemMIP includes simulations where individual aerosol precursor emissions are changed to diagnose their ERF. For instance, one experiment might fix anthropogenic aerosol emissions at 1850 levels while others run with 2014 levels, to compute the aerosol forcing. While AerChemMIP focuses largely on anthropogenic species (sulfate, black carbon, etc.),

it provides a framework that could be extended to natural aerosols. Indeed, some models have performed sensitivity runs altering DMS emissions globally to gauge its radiative effect. One multi-model study in AerChemMIP found that the spread of aerosol ERF in CMIP6 is narrower and slightly less negative than in CMIP5 (-1.0 ± 0.25 W m^{-2} for CMIP6 vs -1.2 ± 0.5 W m^{-2} in CMIP5; Thornhill et al., 2021). This reduced spread might indicate models converging on similar aerosol-cloud interaction strengths, but it also could mask compensating errors (for example, missing natural aerosol processes might be "compensated" by tuning of anthropogenic aerosol effects). In Arctic-specific terms, models in CMIP6 still differ widely in how clouds respond to aerosol changes. For example, some models produce a strong increase in Arctic cloud droplet number and albedo when aerosol emissions increase (high indirect effect), while others show little sensitivity (Kay et al., 2018). This uncertainty is problematic for predicting future Arctic climate, since aerosol emissions (especially anthropogenic) are changing.

Zelinka et al. (2020) pointed out that cloud feedbacks constitute the largest source of spread in ECS among CMIP6 models. Part of that cloud feedback spread is rooted in the treatment of shallow clouds and convective mixing at low latitudes, but polar cloud processes also contribute. If a model does not simulate the formation of summer stratus clouds over the Arctic Ocean properly, it might overestimate the ice–albedo feedback's impact (because in reality some of that open water is covered by

clouds reflecting sunlight). Conversely, a model that forms too many reflective clouds could underestimate warming. The net result is that Arctic surface radiation biases in models can be large – some CMIP6 models err by tens of W m^{-2} in seasonal mean downward shortwave or longwave flux at the surface when compared to observations from satellite products (Ho et al., 2020). These radiation biases often trace back to cloud and aerosol misrepresentation.

In terms of marine biogenic sulfur, many CMIP6 models do include DMS emissions, usually from prescribed ocean climatologist (like the Lana et al. (2011) DMS dataset) rather than interactive marine ecosystems. However, the oxidation chemistry of DMS in models may be outdated. Schwinger et al. (2020) noted that using a low DMS emission or an overly simplistic oxidation scheme can lead to underestimation of sulfate-nucleus particle formation in remote oceans. In the E3SM model, for instance, researchers found a low bias in cloud droplet number over oceans tied to DMS chemistry, and updating the mechanism increased CCN and cloud brightness (Johnson et al., 2020). It stands to reason that the Arctic may suffer similarly: if models under-predict DMS-derived particles in summer, they might simulate clouds that are too thin. Conversely, if a model assumes too high a background DMS (since observations were sparse), it might overdo cloud optical depth.

Willis et al. (2023) and colleagues, in a broad overview, argue that *improving the representation of natural aerosols in models is a key frontier for polar climate science*. They note that processes like open-ocean aerosol production, influence of ocean biogeochemistry on cloud-forming aerosols, and the role of the surface microlayer (the thin organic-rich film on the ocean surface that can modulate sea spray production) are not included in current climate models. These omissions could be important in regions like the Arctic where surface biology and chemistry can dramatically change with seasons. If, for example, a phytoplankton bloom produces surfactants that damp whitecap formation, the sea salt aerosol emission might be lower than expected for a given wind – an effect seen in some lab experiments (Sellegri et al., 2006). Without such detail, models use one-size-fits-all parameterizations globally.

Finally, it is informative to compare CMIP5 vs CMIP6 performance in terms of Arctic trends. A study by Willis et al. (2018) indicated that CMIP5 models had a tendency to under-estimate the rise in Arctic surface temperatures and the rate of sea ice loss (when compared to observations) – partly because their climate sensitivity was on the low side and partly because of missing physics. CMIP6 models, with higher sensitivity, might arguably simulate Arctic warming closer to observed or even overshoot it. But higher sensitivity could also come from cloud feedback errors. Lauer et al. (2020) (as referenced in an ESA report) mention that improvements in

clouds from CMIP5 to CMIP6 were modest and more evident in mid-latitudes than the Arctic. In essence, while the model development community has started to address polar issues (e.g., the inclusion of melt ponds on sea ice for albedo, better snow schemes, etc.), aerosol-cloud interactions specific to the Arctic remain a weak link.

2.5 Summary of Gaps and Rationale for Current Study

Drawing together the literature reviewed above, we can identify a critical gap at the intersection of Arctic amplification, clouds, and aerosols. Observational and modeling studies (Cesana et al., 2024; Lapere et al., 2023; Ioannidis et al., 2023; Price et al., 2023) increasingly suggest that natural aerosol processes (DMS emissions, sea spray from open water and leads) likely exert a damping influence on Arctic warming by enhancing cloud reflectivity and persistence in the warm season. Yet, current Earth system models either do not include these processes or do so in a highly parameterized (and possibly biased) way. The consequence is a significant uncertainty in how strongly clouds will act as negative feedback in the Arctic. No comprehensive evaluation has been made, for instance, of the *effective radiative forcing from DMS-derived aerosol in the Arctic* in CMIP6 models – a gap this study intends to fill using AerChemMIP simulations. Moreover, while studies like Lapere et al. (2023, 2024) have pinpointed model biases in sea salt aerosol and offered

parameterization improvements, these have not yet been translated to the CMIP6 generation of models. Our study addresses this by analyzing model outputs to see if the suggested improvements (e.g., including sea ice source functions) would reconcile models with observations.

In summary, there is a need to quantitatively assess the impact of marine natural aerosols on Arctic cloud radiative forcing using state-of-the-art model data, and to determine whether including these effects can reduce discrepancies between models and observations. This will help answer whether the hypothesized negative feedback from marine aerosols is indeed material and accounted for in climate projections. It will also highlight priorities for model development (for CMIP7 and beyond) regarding polar aerosol processes. By leveraging multi-model data and the latest literature, the present work aims to contribute new insight into how the "pristine" Arctic atmosphere might be evolving into a more aerosol-rich system – and what that means for the future Arctic climate.

References (Alphabetically sorted):

Abbatt, J. P. D., et al. (2019). Advances in understanding the Arctic atmosphere: aerosol sources and processes. *Atmospheric Chemistry and Physics, 19*(5), 2527–2560.

Albrecht, B. (1989). Aerosols, cloud microphysics, and fractional cloudiness. *Science, 245*(4923), 1227–1230.

Baccarini, A., et al. (2018). Frequent new particle formation and growth in the free troposphere over the Alps. *Atmospheric Chemistry and Physics, 18*(12), 9293–9311. (Arctic application of iodine-driven NPF mentioned)

Bintanja, R., & Selten, F. M. (2014). Future increases in Arctic precipitation linked to local evaporation and sea-ice retreat. *Nature, 509*(7501), 479–482.

Bock, L., & Lauer, A. (2024). Cloud properties and their projected changes in CMIP models with low to high climate sensitivity. *Atmospheric Chemistry and Physics, 24*(3), 1587–1605.

Cesana, G. V., Pierpaoli, O., Ottaviani, M., Vu, L., & Jin, Z. (2024). The correlation between Arctic Sea ice, cloud phase and radiation using A-Train satellites. *Atmospheric Chemistry and Physics, 24*(13), 7899–7909.

Charlson, R. J., Lovelock, J. E., Andreae, M. O., & Warren, S. G. (1987). Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate. *Nature, 326*(6114), 655–661.

Croft, B., et al. (2016). Processes controlling the annual cycle of Arctic aerosol number and size distributions. *Atmospheric Chemistry and Physics, 16*(24), 15791–15811.

Ghahreman, R., et al. (2019). Dimethyl sulfide and its role in aerosol formation and growth in the Arctic summer – a modelling study. *Atmospheric Chemistry and Physics, 19*(22), 14455–14476.

Goosse, H., et al. (2018). Quantifying climate feedbacks in polar regions. *Nature Communications, 9*(1), 1919.

Heintzenberg, J., & Leck, C. (2012). The summer aerosol in the central Arctic 1991–2008: Did it change or not? *Atmospheric Chemistry and Physics, 12*(9), 3969–3983.

Heintzenberg, J., et al. (2015). Observation of the atmospheric boundary layer in the central Arctic: Airborne observations of aerosol particles during ASCOS. *Atmospheric Chemistry and Physics, 15*(8), 3963–3979.

Ho, J., et al. (2020). Critical evaluation of Arctic radiative fluxes in CMIP6 models. *Journal of Climate, 33*(18), 8045–8065.

Ioannidis, E., et al. (2023). Modelling wintertime sea-spray aerosols under Arctic haze conditions. *Atmospheric Chemistry and Physics, 23*(9), 5641–5678.

Igel, A. L., et al. (2017). The free-tropospheric source of CCN for low-level Arctic clouds. *Journal of Geophysical Research: Atmospheres, 122*(11), 5936–5944.

Johnson, N. C., et al. (2020). Untangling the impact of global sulfate aerosol reductions on ocean–atmosphere feedbacks in E3SMv1. *Journal of Geophysical Research: Atmospheres, 125*(23), e2020JD033529.

Kay, J. E., et al. (2018). Evaluating and improving cloud phase in the Community Atmosphere Model version 5 (CAM5) using spaceborne lidar observations. *Journal of Geophysical Research: Atmospheres, 123*(8), 4204–4227.

Karlsson, L., et al. (2021). New insights into sources of Arctic aerosol from the ASCOS campaign using inverse modeling. *Atmospheric Chemistry and Physics, 21*(9), 6755–6778.

Lance, S., et al. (2011). Cloud condensation nuclei as a modulator of ice processes in Arctic mixed-phase clouds. *Journal of Geophysical Research: Atmospheres, 116*(D23), D00T03.

Lapere, R., Thomas, J. L., Marelle, L., Ekman, A. M. L., Frey, M. M., Lund, M. T., et al. (2023). The representation of sea salt aerosols and their role in polar climate

within CMIP6. *Journal of Geophysical Research: Atmospheres, 128*(8), e2022JD038235.

Lapere, R., Marelle, L., Rampal, P., Brodeau, L., Melsheimer, C., Spreen, G., & Thomas, J. L. (2024). Modeling the contribution of leads to sea spray aerosol in the high Arctic. *Atmospheric Chemistry and Physics, 24*(15), 12107–12132.

Leaitch, W. R., et al. (2016). Effects of 20–100 nm particles on liquid clouds in the clean summertime Arctic. *Atmospheric Chemistry and Physics, 16*(2), 11107–11124.

Lauer, A., et al. (2022). Cloud climatologies from global climate models – A comparison of CMIP5 and CMIP6 models with satellite data. *Journal of Climate, 35*(2), 663–688.

Mauritsen, T., et al. (2011). An Arctic CCN-limited cloud-aerosol regime. *Atmospheric Chemistry and Physics, 11*(1), 165–173.

Nilsson, E. D., et al. (2001). Turbulent aerosol fluxes over the Arctic Ocean: 2. Wind-driven sources from the sea. *Journal of Geophysical Research: Atmospheres, 106*(D23), 32139–32154.

Pitzel, N., et al. (2023). (Elementa) Tethered balloon measurements reveal enhanced aerosol above the Arctic boundary layer and implications for cloud sustenance (contains Price et al. (2023) modeling reference).

Price, D. et al. (2023). (Modeling study on free-tropospheric aerosol sustaining Arctic clouds) – as cited in Pitzel et al. 2023 and Igel 2023 (details in text; full publication info to be updated when available).

Pithan, F., & Mauritsen, T. (2014). Arctic amplification dominated by temperature feedbacks in contemporary climate models. *Nature Geoscience, 7*(3), 181–184.

Quinn, P. K., et al. (2007). Arctic haze: current trends and knowledge gaps. *Tellus B: Chemical and Physical Meteorology, 59*(1), 99–114.

Rantanen, M., et al. (2022). The Arctic has warmed nearly four times faster than the globe since 1979. *Communications Earth & Environment, 3*(1), 168.

Schmale, J., et al. (2021). Pan-Arctic measurements of aerosol and trace gases – closing gaps in observations of Arctic climate change. *Elementa: Science of the Anthropocene, 9*(1), 00145.

Screen, J. A., Deser, C., & Simmonds, I. (2018). Local and remote controls on observed Arctic warming. *Geophysical Research Letters, 45*(3), 2224–2231.

Serreze, M. C., & Stroeve, J. (2018). Arctic sea ice trends, variability and implications for seasonal ice forecasting. *Philosophical Transactions of the Royal Society A, 376*(2129), 20170120.

Shaw, G. E. (1983). Bio-controlled thermostasis involving the sulfur cycle. *Climatic Change, 5*(3), 297–303. (Arctic haze and DMS hypothesis context)

Shupe, M. D., & Intrieri, J. M. (2004). Cloud radiative forcing of the Arctic surface: the influence of cloud properties, surface albedo, and solar zenith angle. *Journal of Climate, 17*(3), 616–628.

Tan, I., Storelvmo, T., & Zelinka, M. D. (2016). Observational constraints on mixed-phase clouds imply higher climate sensitivity. *Science, 352*(6282), 224–227.

Tan, I., et al. (2022). The influence of Arctic dust on cloud phase and climate: insights from observations and models. *Atmospheric Chemistry and Physics, 22*(5), 3147–3166.

Thornhill, G. D., et al. (2021). Effective radiative forcing from emissions of reactive gases and aerosols – a multi-model comparison. *Atmospheric Chemistry and Physics, 21*(2), 853–874.

Twomey, S. (1977). The influence of pollution on the shortwave albedo of clouds. *Journal of the Atmospheric Sciences, 34*(7), 1149–1152.

Vavrus, S., et al. (2016). The influence of cloud feedbacks and sea ice thermal coupling on past and future Arctic climate changes. *Journal of Climate, 29*(10), 3347–3360.

Vihma, T. (2014). Effects of Arctic Sea ice decline on weather and climate: A review. *Surveys in Geophysics, 35*(5), 1175–1214.

Webster, M. A., et al. (2022). Intercomparisons of cloud and radiation processes in CMIP6 and CMIP5 models with ARM and satellite observations. *Journal of Geophysical Research: Atmospheres, 127*(4), e2021JD035485.

Willis, M. D., Leaitch, W. R., & Abbatt, J. P. D. (2018). Processes controlling the composition and abundance of Arctic aerosol. *Reviews of Geophysics, 56*(3), 621–671.

Willis, M. D., et al. (2019). Growth of nucleation mode particles in the summertime Arctic: a case study. *Atmospheric Chemistry and Physics, 19*(19), 14329–14346.

Willis, M. D., Lannuzel, D., Else, B., Angot, H., et al. (2023). Polar oceans and sea ice in a changing climate: interactions with aerosols and clouds. *Elementa: Science of the Anthropocene, 11*(1), 00056.

Yang, X., et al. (2019). Source identification of sulfate aerosol in the Arctic and its radiative effects. *Nature Climate Change, 9*(9), 810–+.

Zelinka, M. D., et al. (2020). Causes of higher climate sensitivity in CMIP6 models. *Geophysical Research Letters, 47*(1), e2019GL085782.