Thesis for The Degree of Licentiate of Engineering

Modeling aerosol-cloud interactions in the Arctic

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Göteborg, Sweden, 2022

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Cover: Mixed-phase clouds over Arctic sea ice. Image taken by Luisa Ickes.

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Modeling aerosol-cloud interactions in the Arctic

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Abstract

Clouds have a large impact on Earth's energy balance, especially in the Arctic. Through their warming or cooling effect on the surface, clouds can play a critical role in the onset of melting and freezing of Arctic sea ice, which itself has a large effect on energy and moisture fluxes between the ocean and the atmosphere. One of the factors that determine whether a cloud has a net warming or cooling effect is its microphysical structure determined by the phase of the hydrometeors that the cloud consists off. Mixed-phase stratiform clouds, consisting of both water droplets and ice crystals, often occur in the Arctic between mid-spring and mid-fall. To be able to simulate the Arctic climate, it is crucial that models capture Arctic mixed-phase stratiform clouds (AMPS) and the apportionment between liquid and frozen hydrometeors in these clouds. A good representation of ice nucleation is the necessary first step for accurate modeling of cloud ice. Ice nucleation in mixed-phase clouds occurs heterogeneously with the requirement of ice nucleating particles (INP). This work presents a new heterogeneous freezing parameterization that was tested in a large-eddy simulation of AMPS. Different to other parameterization schemes, this parameterization does not require knowledge about the aerosol concentration and characteristics (type, size, etc.). Instead the parameterization is based on the observation that the frequency of INP concentrations at a specific temperature follows a log-normal distribution and randomly draws INP concentrations from this distribution at the present temperature. It is shown that the new parameterization results in reasonable amounts of cloud ice and that the random drawing of INP concentrations is an important aspect to be investigated when it comes to cloud ice formation.

Keywords

Mixed-phase clouds, heterogeneous freezing, parameterization, ice nucleating particles, LES-modeling, Arctic.

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Hannah Frostenberg
Göteborg, November 2022

List of Publications

Appended publications

This thesis is based on the following appended papers:

Paper 1. Hannah C. Frostenberg, André Welti, Mikael Luhr, Julien Savre, Erik S. Thomson, Luisa Ickes, *The Chance of Freezing – Parameterizing temperature dependent freezing including randomness of INP concentrations*
Atmospheric Chemistry and Physics Discussions. Submitted, under review. DOI: 10.5194/acp-2022-696

Contents

Part I

Introductory chapters

Chapter 1

Clouds in the climate system

Clouds are ubiquitous in most locations on Earth, apart from the regions of climatological subsidence (see Fig. 1.1). They form when moist air rises and reaches saturation, upon which the water vapor in the air condenses to liquid droplets or deposits to ice crystals.

The most important direct effect of clouds for humans is their role in the water cycle, since they transport moisture and energy, and are the source of precipitation. However, clouds also interact with both the incoming shortwave (solar) radiation and outgoing longwave (terrestrial) radiation. Both

Figure 1.1: Earth, picture by NASA's Deep Space Climate Observatory (DSCOVR) satellite.
Image Credit: NASA

components, clouds' contribution to the water cycle and energy balance, make them crucial elements for Earth's climate. Additionally, the surface of cloud droplets and ice crystals can catalyze chemical reactions in the atmosphere that, e.g., form ozone-depleting substances in the polar stratosphere, which caused the Antarctic ozone hole (e.g., Solomon, 1988). Even though clouds are important for Earth's climate, there are still large uncertainties of how clouds themselves affect and will be affected by global climate and climate change (IPCC, 2021). This is because there are many factors influencing cloud properties, which in turn lead to different climatic effects as will be explained in the following sections and chapters. The uncertainties in clouds are the most important source for our uncertainty in how future climate will respond to the anthropogenic impact on global climate.

1.1 Classifications of clouds

Clouds can be distinguished by the phase of the cloud particles (so-called hydrometeors) they are made up from: **warm clouds** contain solely liquid cloud droplets and rain drops, **ice clouds** consist of only frozen hydrometeors like ice crystals and snow flakes, and **mixed-phase clouds** contain both liquid and frozen hydrometeors. Apart from these definitions, meteorologists have developed an elaborate system of cloud classification based on their appearance, following the system of genera, species and varieties used in, e.g., botany (WMO, 2017). The main categories are "cirrus", "stratus" and "cumulus".
Cirrus clouds are high clouds consisting of ice crystals, which makes them almost transparent to sunlight and gives them a feather-like appearance (Fig. 1.2 a). They often have blurry edges due to ice crystals slowly sedimenting out of the cloud (Penner et al., 1999). These sedimenting ice crystals sublimate in drier air and thus do not reach the ground. Cirrus clouds have a typical lifetime of several hours and are formed by lifting of air until the water vapor deposits. **Stratus** clouds are much larger in their horizontal dimension than in their vertical dimension and often form a closed layer (Fig. 1.2 b). They can consist of liquid or frozen hydrometeors, as well as a mixture of both and can produce light precipitation. Their lifetime ranges from 6 to 12 hours (Cotton et al., 2010). Stratiform clouds typically form from large-scale lifting of layers of air until the air reaches saturation and water vapor condenses (Lohmann et al., 2016). In contrast, **cumulus** clouds form by convective motions, when air warmed by the surface rises as air pockets up to saturation. They have a defined bottom and fluffy, often well-defined outlines (Fig. 1.2 c). As stratus clouds, they can consist of hydrometeors of any phase, but their majority contains liquid droplets that cause the well-defined edges. Cumulus clouds often have a short lifetime of less than one hour (Cotton et al., 2010) and they might produce rain or snow showers.

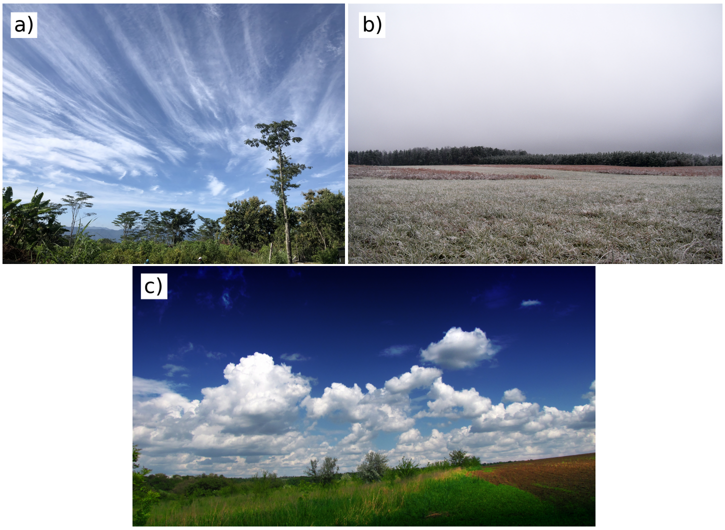

Figure 1.2: Different cloud types: a) cirrus, b) stratus and c) cumulus. Image Credit: Wikimedia Commons

1.2 Clouds and the global energy balance

Clouds interact with both outgoing longwave and incoming shortwave radiation. Figure 1.3 illustrates that the effect on radiation depends strongly on the cloud's height, temperature and the phase of its hydrometeors. Low, and thus mainly warm clouds, consist primarily of liquid droplets. Cloud albedo (shortwave reflectivity) is proportional to the amount of liquid water and the number of liquid droplets (Seinfeld and Pandis, 2016), which makes warm clouds highly reflective to incoming shortwave radiation: they can have an albedo of up to 90% (Hartmann, 1994). This means that low clouds are highly reflective to incoming shortwave radiation. Low-level clouds exert only a small greenhouse effect, because most of the longwave radiation emitted in the lower atmosphere (and thus also by low clouds) will be absorbed again in the lower atmosphere instead of escaping to space. The overall effect for low-level clouds thus is a cooling effect (Matus and L'Ecuyer, 2017). High cirrus clouds are almost transparent for shortwave radiation with typical albedos below 0.3 (Fu and Liou, 1993), since they consist of no or very little liquid water and ice crystals are much less reflective than liquid droplets. Ice clouds have a considerable greenhouse effect, because they reside in altitudes with low temperatures: Seen from space, a high cloud emits much less longwave radiation due to its low temperature compared to a cloud-free atmosphere emitting from levels with higher temperature. This causes a strong greenhouse effect because the excessive radiation is kept within the atmosphere. Overall, cirrus clouds have a net warming effect (Matus and L'Ecuyer, 2017).
Another important aspect of a cloud's radiative effect is the albedo of the underlying surface. The lower the surface albedo is, the higher the cloud's

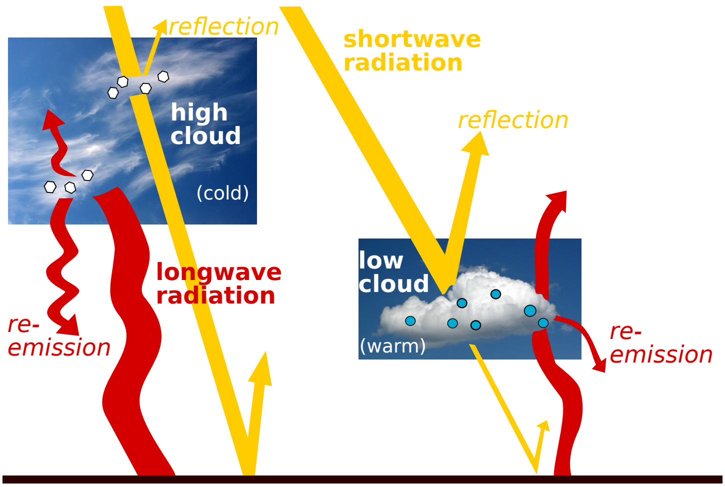

Figure 1.3: Clouds' impact on radiation.
Image Credit for photos: Luisa Ickes; James St. John, CC BY 2.0, via Wikimedia Commons

shortwave effect becomes. This means that low-level clouds have a larger shortwave effect over dark surfaces like open ocean than over, e.g., highly reflective ice or snow. Indeed, low-level clouds in polar regions have a net warming effect through most of the year due to the high surface albedo and large solar zenith angles (Shupe and Intrieri, 2004).
Apart from affecting radiation, clouds are part of another energy redistribution between the surface and the atmosphere, as well as between different locations: latent heat. When liquid water at Earth's surface is being evaporated, the latent heat of vaporization of 2 500 kJ/kg has to be exerted. Upon condensation in a cloud, this latent heat is released into the atmosphere. Since evaporation and condensation can occur at different geographical locations, latent heat is part of the global energy redistribution.

1.3 Clouds in a changing climate

By the sheer warming of the atmosphere through global warming we can expect changes in clouds. The isotherm of the melting point will shift upwards in the atmosphere, which elevates the part of the atmosphere where frozen hydrometeors can exist. This will affect radiative properties of clouds, since the partitioning between ice and liquid in clouds will shift towards more liquid, which makes clouds more reflective for sunlight (see Sect. 1.2). Additionally, circulation patterns in a future climate are expected to change (e.g., Demuzere et al., 2009). This will lead to different spatial distributions of clouds in the future, which can have an effect on precipitation patterns, especially where monsoon regions are affected (e.g., Sandeep et al., 2018). Also regarding clouds' radiative effect, their location matters if they move over a surface with different albedo or to different latitudes where the strength of the solar radiation differs. Changes in cloud type and location caused by the changing climate lead to further perturbations of climate. This mechanism is called cloud feedback and it can be positive (amplifying the initial perturbation, e.g., global warming) or negative (dampening the initial perturbation and stabilizing the climate). The overall strength of the cloud feedback on Earth's climate is one of the largest uncertainties in global climate modeling, even though progress has been made (IPCC, 2021). The latest IPCC report AR6 (IPCC, 2021) gives a best estimate of net cloud feedback as 0.42 W $m^{-2}{}^\circ C^{-1}$ with the very likely range of -0.10 to 0.94 W $m^{-2}{}^\circ C^{-1}$. The best estimate is a positive feedback and leads to an additional radiative forcing of 0.84 W m^{-2} for a warming of 2°C.

Chapter 2

Cloud microphysics

Clouds are made of different hydrometeor types: cloud droplets and rain drops, ice crystals, snow flakes, graupel and hail. Cloud microphysics covers the formation of these hydrometeors and their growth into precipitation-size particles (Pruppacher and Klett, 1997). The size, phase and shape of a cloud's hydrometeors affect the cloud's macrophysical properties like its albedo, lifetime, spatial extent and precipitation. To estimate the microphysical structure of a cloud, the most relevant variables are number concentrations of the different hydrometeors and their mass (often represented by mixing ratio). Ideally, these variables are known for all occurring sizes of a hydrometeor species, but often only bulk values (averaged over all hydrometeors of a certain category) can be observed or modeled.

2.1 Latent heat

When a phase transition occurs, e.g., water vapor condensing to a droplet, latent heat is released to the environment if the energetic state of the second phase is lower than that of the first state (as in the case of condensing water vapor). Otherwise, energy is needed to transfer the matter to a phase with a higher energetic state (e.g., vaporizing a droplet). The latent heat released by or required for the phase transition is absorbed or provided by the environment, which means that the temperature of the water mixture does not change during the phase transition, while the temperature of the surrounding air does.

2.2 Activation of cloud droplets

In order for water vapor to condense, the air must be supersaturated with it. This means that the partial pressure of water vapor is larger than the equilibrium vapor pressure and the air cannot "hold" the water vapor in gaseous form anymore. However, supersaturations of approximately 500% would be required for water droplets to form homogeneously, i.e., without an assisting aerosol particle (Lohmann et al., 2016). The reason is that for

a small amount of water molecules, the cost of building a surface between the liquid droplet and the surrounding air is exceeding the gain of energy by the phase transition from gaseous to liquid. The fewer water molecules the newly-formed droplet contains (and thus the smaller its radius is), the larger the cost of surface building. This means that for water droplets with a finite radius, the equilibrium vapor pressure is larger than over a flat water surface and even larger for smaller droplets (Kelvin effect, see dashed line in Fig. 2.1). Hydrophilic soluble aerosols can serve as cloud condensation nuclei (CCN) by adsorbing water molecules (i.e., they cause the phase transition to liquid) already below a relative humidity of 100%. The particle grows further by accumulating water molecules below saturation (hygroscopic growth) and at some point, the aerosol is dissolved in the formed cloud droplet. The molecules of the former CCN in the droplet lower the equilibrium vapor pressure around the deliquesced particle (Raoult's law, dash-dotted lines in Fig. 2.1).
Köhler theory combines the Kelvin effect (enhanced saturation pressure over curved droplets) and Raoult's law (reduced saturation pressure over water solutions) to describe the heterogeneous formation of cloud droplets (solid lines in Fig. 2.1). For small droplet sizes, Raoult's effect dominates and the droplet grows even below supersaturation. The more the droplet grows, the larger the equilibrium vapor pressure over the solution droplet becomes until it reaches a maximum (activation saturation ratio S_{act} at critical droplet radius or activation radius r_{act}) and the droplet grows spontaneously. The larger

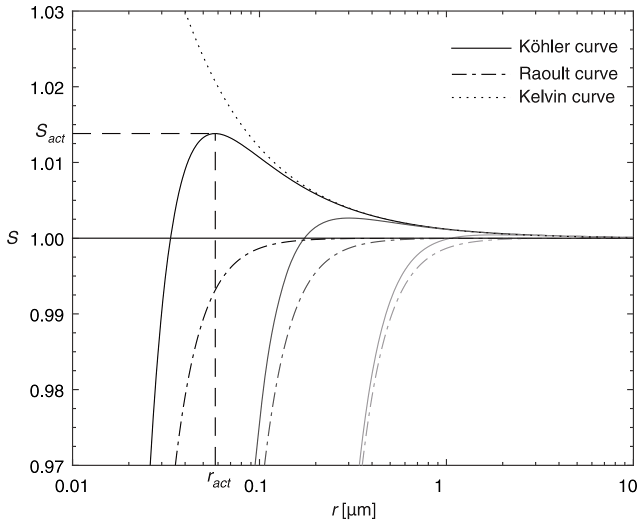

Figure 2.1: Köhler and Raoult curves of three NaCl particles with different dry radii (left to right: 0.01 µm, 0.03 µm and 0.1 µm). Kelvin curve is the same for all particles. S is size-dependent saturation ratio of the solution droplet: equilibrium vapor pressure over the solution droplet with radius r divided by saturation vapor pressure over a flat surface of pure water. Adapted from Lohmann et al. (2016).
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the droplet gets, the more important the Kelvin effect becomes relative to the Raoult effect (see Fig. 2.1). In summary, soluble aerosols are required to form cloud droplets since they considerably lower the saturation ratio necessary for water vapor to condense to liquid droplets.

2.3 Ice nucleation

Only at temperatures below approximately -38°C can supercooled liquid droplets in the atmosphere freeze **homogeneously**, i.e., without an aerosol particle inducing the phase change. At higher temperatures, ice nucleating particles (INPs) are crucial for ice initiation (**heterogeneous** ice nucleation). The different pathways to nucleate atmospheric ice crystals, referred to as modes, are:

- Homogeneous ice nucleation: freezing of a cloud or solution droplet, without interaction with an INP.
- Immersion freezing: an INP is suspended in a supercooled cloud droplet and at a specific temperature, which depends on properties of the INP, the INP induces freezing of the cloud droplet.
- Contact freezing: an INP collides with a supercooled droplet and initiates its freezing immediately.
- Deposition ice nucleation: water vapor deposites directly to an INP.
- Condensation freezing: water first condenses on the INP at sub-zero temperatures and immediately freezes.

Numerous observational studies of mixed-phase clouds have shown that liquid cloud droplets exist before ice nucleation occurs (e.g., Ansmann et al., 2009; de Boer et al., 2011; Westbrook and Illingworth, 2011), which makes immersion and contact freezing the most relevant modes in mixed-phase clouds. Contact freezing is further limited by the probability for collisions that are necessary between the supercooled droplet and interstitial aerosol (non-activated aerosol).

2.3.1 Heterogeneous ice nucleation

Similar to the phase change of a condensing droplet (Sect. 2.2), an energy barrier has to be overcome in order to form ice from supercooled liquid water. The barrier is caused by the counteracting energy required for building a surface between the supercooled water and the growing ice, and the gain in energy when a water molecule changes from the liquid to the solid phase. Analogous to a CCN, an INP lowers the energy barrier by providing a surface onto which a water molecule can attach and change its phase more easily (Whale, 2018). Different from the activation of cloud droplets, ice nucleation has a strong dependency on temperature. However, in experiments it has also been found that with time, more droplets will freeze at the same conditions. This has lead to two views on heterogeneous ice nucleation: that it either is a **deterministic** or a **stochastic** process. The deterministic concept postulates that each individual INP has a specific temperature or saturation with respect to ice at which ice will begin to nucleate on its surface, or more precisely on so-called *active sites* of the INP. This means that there are “good” and “bad” INPs for triggering freezing. If a freezing experiment is repeated with the same set of droplets and INPs, the same droplets will freeze at the exact same conditions as in a previous experiment. Time itself has no impact on the amount of frozen droplets. In the stochastic view on the other hand, there is an inherent random aspect to heterogeneous ice nucleation and it is not possible to assign an INP certain conditions at which ice always will form on its surface. Instead, when keeping temperature and ice saturation at the same level, more droplets will freeze over time. Repeating a freezing experiment with the same set of droplets and INPs will lead to roughly the same amount of droplets freezing, but it is not necessarily the same droplets that freeze at the same conditions as in a previous experiment (Lohmann et al., 2016). According to a review of laboratory experiments by Vali (2014), surface characteristics and their temperature dependence of INPs have a larger influence on heterogeneous immersion freezing than time. This indicates that the deterministic aspect of heterogeneous freezing dominates over the stochastic aspect, but under certain conditions the time dependence of freezing still has to be taken into account. Heterogeneous freezing parameterizations use either of these assumptions. Examples of deterministic parameterizations are the parameterization by Niemand et al. (2012) which depend on the surface area of dust aerosol, the parameterization by DeMott et al. (2010) depending on aerosols with diameter larger than 0.5 μm and the one by Phillips et al. (2008) that describes immersion, contact and deposition nucleation on three types of aerosol. In these parameterizations, a determined number of nucleation events occur at a specific temperature that depends on the aerosol size, type or number of active sites. Parameterizations applying the stochastic view use classical nucleation theory, where a nucleation rate is calculated based on aerosol size and characteristics. Examples of stochastic parameterizations are the one by Hoose et al. (2010) which represents immersion, contact and deposition nucleation on five aerosol types and by Ickes et al. (2017) describing immersion freezing on mineral dust particles.

2.3.2 Ice nucleating particles

Pruppacher and Klett (1997) list a number of criteria that an INP has to fulfill, namely insolubility, size, a chemical bond requirement, crystallographic requirement and active site requirement. However, Murray et al. (2012) summarize several studies that disconfirm that all these requirements need to be met by INPs. It is clear however, that a solid surface is a prerequisite for aerosols to be able to act as INP. Despite that, INP characteristics can differ depending on the aerosol type (for some, crystallography might be a dominating factor for the freezing characteristics, for others active sites etc.). The requirement of specific properties means that ice nucleating particles are a very small subset

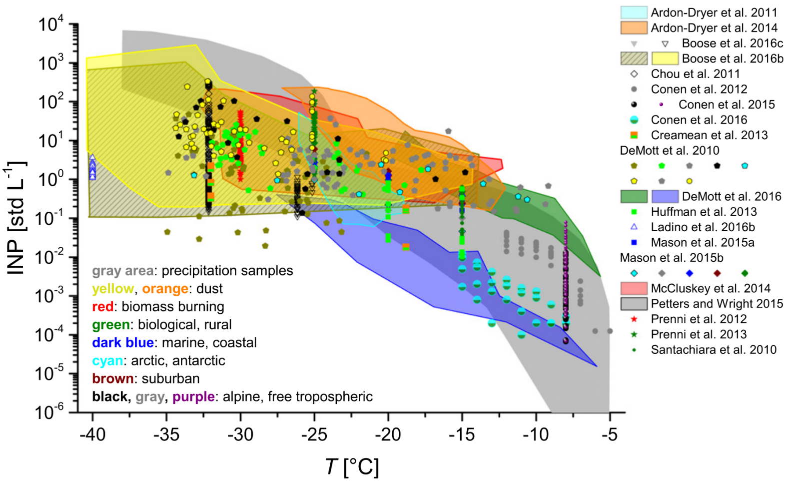

Figure 2.2: Compilation of INP observations, from Kanji et al. (2017).

of atmospheric aerosols: only about 10^{-6} to 10^{-5} of atmospheric aerosols can act as INPs (Lohmann et al., 2016). Total aerosol number concentrations can amount to between 100 and 100 000 cm^{-3} (Koutsenogii and Jaenicke, 1994). Different aerosol types can act as INPs (Fig. 2.2): mineral dust particles are the most extensively investigated INPs (the review of INP laboratory experiments by Hoose and Möhler (2012) lists 61 studies on mineral dusts, followed by 35 bioaerosol studies), but mineral dust particles are active at lower temperatures below ca. -15°C. At higher temperatures, it is mainly biological aerosol that has been found to initiate ice nucleation. This can be proteins produced by bacteria, as well as (parts of) pollen or other plant-material. Oceans are another considerable INP source, producing both sea salt aerosol and secondary organic particles.

Measuring ambient INP concentrations

Two main ways of measuring ambient INP concentrations (INPCs) exist. One is collecting aerosol on filters, and subsequently immersing them in a liquid that makes growing ice crystals easily detectable (e.g., Bigg et al., 1963; Wex et al., 2019). The immersed particles are cooled down and the number of frozen droplets or ice crystals is being registered at the given temperatures. This method requires the sampling of a large enough air volume in order to collect a sufficient number of INPs, which means that the measurements are done on rather coarse time resolution of typically one day. The filter method is mainly being used for temperatures down to -25°C, because unavoidable impurities in the liquid cause freezing below approximately this threshold.

Using cloud chambers like continuous flow diffusion chambers (CFDCs) is another method of measuring ambient INPC (e.g., Rogers, 1988; Garimella et al., 2016). A continuous stream of air containing aerosol is analysed in-situ. The air stream is directed through a cloud chamber, where the walls are ice-covered and can be heated differentially in order to yield a specific temperature and supersaturation inside the chamber. Ice crystals that have grown on INPs in the flow are detected by optical instruments at the end of the chamber. To ensure that activated liquid droplets are not mistaken for ice crystals, the flow passes an evaporation section before counting, where the vapor pressure is kept at ice saturation. Depending on the specific instrument's setup, a temperature range of about -15 to -50°C can be covered by CFDCs. This is limited by the optical detection instruments, since a minimum fraction of droplets has to be frozen in order to be detected. However, the number of frozen droplets in turn depends on the effectiveness of the INPs that are measured.

The characteristic distribution of INP concentrations

Fig. 2.2 shows the cumulative INP concentration depending on temperature. The exponential dependency of INP concentrations on temperature is caused by the probability that a small cluster of water molecules forms an initial small ice cluster in the supercooled droplet triggering the macroscopic phase change. It results from the dependency of the molecules' speed on temperature which is an exponential relationship. The lower the temperature, the slower the molecules and the higher the probability that water molecules will bind together.

When measuring the relative distribution of ambient INPCs at a specified temperature (basically along a vertical line in Fig. 2.2, not shown), it has been found that the distribution often resembles a log-normal distribution (e.g., Isaac and Douglas, 1971; Welti et al., 2018). This indicates that the INP population had been subject to substantial dilution since their emission into the atmosphere, and thus that INP sources are far away from measurement locations (Ott, 1990).

2.4 Growth and sinks of hydrometeors

Newly formed cloud droplets or ice crystals are so light that they are held aloft by the vertical winds in the cloud. To become a precipitation hydrometeor and leave the cloud, they need to grow considerably. For example, a typical cloud droplet has a radius of 5 μm while a typical raindrop has a radius of 1 mm (Lohmann et al., 2016). This means that a cloud droplet's mass needs to increase by a factor of 7 orders of magnitude to become a raindrop falling towards the ground.

2.4.1 Diffusional growth by water vapor, evaporation and sublimation

After liquid droplets have grown up to their activation radius (see Sect. 2.2), they grow spontaneously by diffusion and condensation of water vapor, as long as the environment is supersaturated. The same applies to ice crystals, if

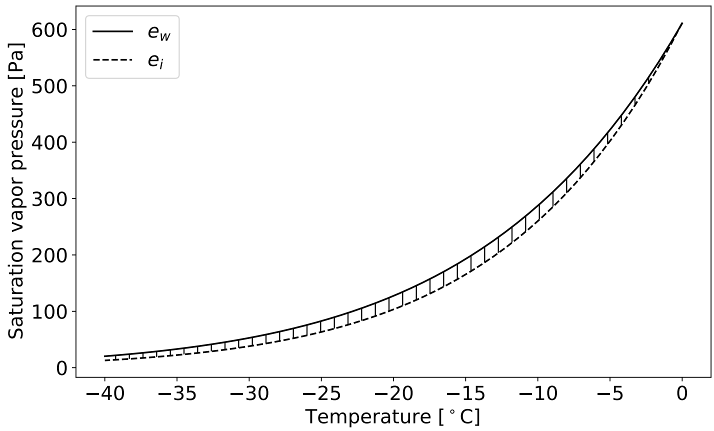

Figure 2.3: Saturation pressures over water (e_w, solid) and ice (e_i, dashed). Area between where WBF process is active is shaded.

the vapor pressure is above saturation with respect to ice. If vapor pressure falls below the respective phase's saturation pressure, liquid droplets begin to evaporate and ice crystals to sublimate. An important aspect of cloud microphysics is that the saturation ratio with respect to ice is always higher than that with respect to water. This means that at a vapor pressure between saturation over water and saturation over ice and a co-existence of liquid and frozen hydrometeors, the frozen particles will grow at the expense of evaporating liquid droplets (Wegener-Bergeron-Findeisen, WBF, process, see Fig. 2.3).

2.4.2 Collection processes

Through diffusional growth alone, hydrometeors would not grow to precipitation-size particles in relevant atmospheric time spans. The collision and collection of different hydrometeors is a much more efficient process for hydrometeors to grow. Collection processes are most efficient when the hydrometeors are of very different sizes (since this means that they have varying fall velocities which increases the collision probability), and if the conditions in the cloud are more turbulent (since the hydrometeors then can have an additional varying velocity component).
The following collection processes are possible in mixed-phase clouds:

- Collision-coalescence: liquid droplets colliding and forming a larger liquid droplet.

- Aggregation: ice crystals collide and stick together forming an ice crystal aggregate.

- Riming: liquid droplets freezing onto a frozen hydrometeor forming graupel or hail.

Note that not every collision of two frozen hydrometeors necessarily leads to a new hydrometeor. Depending on the coalescence (liquid droplets) or sticking efficiency (ice crystals) of both hydrometeors, they might not coalesce or simply bounce off each other. In the mid-latitudes, both over land and oceans, most of the rain falls from clouds that contain ice (Mülmenstädt et al., 2015). They additionally found that almost everywhere over the continents, less than 5% of the rain originates from warm clouds. This indicates that collisions involving frozen hydrometeors are crucial for most precipitation over land to form.

2.4.3 Melting

If temperature rises above the freezing point, ice crystals and snow flakes begin to melt. This does not immediately change the hydrometeor's mass (although the saturation pressure of water is higher than for ice, which means that evaporation is more likely to occur from liquid than frozen hydrometeors), but other properties like its interaction with radiation or the probability to collide and stick to another hydrometeor (see Sect. 2.4.2). Additionally, this phase transition leads to the uptake of latent heat, as all phase transitions to higher energetic states (see Sect. 2.1).

2.4.4 Precipitation

The most efficient sink for a cloud is its hydrometeors falling to the ground as precipitation. When a hydrometeor's fall speed is larger than the updraft that holds it within the cloud, it falls towards the ground. If the air below the cloud is very dry, the hydrometeor might evaporate or sublimate before reaching the surface, supplying moisture to that altitude. Analogously, falling frozen hydrometeors melt if the air below the cloud is above the freezing point, and at the surface they might be rain instead.

2.5 Secondary ice processes

Observations of ice crystal number concentrations in clouds above -38°C often exceed INP concentrations by several orders of magnitude (e.g., Hobbs and Rangno, 1985; Hobbs and Rangno, 1998). Several mechanisms have been described that enhance ice crystal number without ice nucleation, so-called secondary ice processes:

- Collisional breakup (e.g., Vardiman (1978)): colliding ice crystals lead to fragments breaking off of one of the colliding crystals.
- Drop shattering (e.g., Lauber et al. (2018)): while a super-cooled droplet is freezing from the outer-most layer, pressure builds up inside the droplet. This can lead to shattering of the droplet and the already formed frozen surface, producing small ice fragments.
- Rime splintering or Hallett-Mossop process (e.g., Hallett and Mossop, 1974; Scott and Hobbs, 1977): during riming of a super-cooled droplet on

a large ice particle (snow flake, graupel or large frozen drop), secondary ice splinters form due to the pressure build up as in drop shattering. This process is assumed to be active at temperatures between -8 and -3°C.

2.6 Aerosol-cloud interactions

As we have seen in this chapter, aerosols are crucial for clouds to form and develop. They are necessary for cloud droplet formation and for nucleating ice above ca. -38°C. This means that the abundance and type of aerosols determine some characteristics of clouds, most importantly the amount of droplets and ice. Let's compare a cloud that forms over land versus one that forms above sea as an example: there will be more aerosols (total number) in the cloud formed above the continental location compared to the marine location, because there are more particle sources above land. If we assume that the same amount of water is available to condense onto the aerosols in both clouds, the marine cloud will consist of fewer and larger droplets than the continental cloud. A cloud's albedo is proportional to the number of droplets, which means that the continental cloud will be more reflective than the marine cloud and have a larger cooling effect (Twomey, 1974). In order to grow to precipitation-size hydrometeors, cloud particles need to collide and stick together. This is least efficient if hydrometeors of similar sizes exist in the cloud, especially when they are small (see Sect. 2.4.2). The precipitation-formation mechanism will be much more efficient for the marine cloud than for the continental cloud in the example above, because the droplets in the continental cloud are smaller than in the marine cloud. Since precipitation is the most efficient process to dissolve a cloud, the continental cloud will persist longer than the marine cloud and influence radiation for a longer time, adding an additional cooling effect (Albrecht, 1989). To summarize, aerosols have a large impact on clouds, including their lifetime and radiative effect.

Chapter 3

Arctic climate and clouds

3.1 Polar climate

Both polar environments are characterized by the extreme differences in solar insulation throughout the year: during polar summer, the sun never sets while it does not rise above the horizon during polar winter. Additionally, the poles are covered with ice and snow which leads to high reflectivities - apart from open leads or entirely open ocean that have very low albedo. In contrast to Antarctica, which is a continent, the Arctic is an ocean surrounded by the large landmasses of North America and Eurasia. Due to this geographical difference, the respective ice masses are of different nature. Antarctica is covered by a continental ice sheet surrounded by sea ice, while in the Arctic, only Greenland is covered by an ice sheet and the remaining Arctic ice is made up of sea ice. Since surface temperatures are generally low at high latitudes, the absolute water vapor content is also low (due to the temperature dependency of the saturation vapor pressure, e.g., Pruppacher and Klett, 1997). Relative humidity can be high above open ocean areas, where water evaporates from the warmer ocean, but is generally low in the polar atmosphere. Apart from open oceans or open land areas in the Arctic, there are no relevant aerosol sources in the polar environments. Large amounts of warm air, moisture and aerosol are transferred to the poles via meridional transport from lower latitudes. Another consequence of the low polar temperatures is a low tropopause height between ca. 8 and 12 km. Above the tropopause in the stratosphere, an additional distinct feature of polar climate exists: the polar vortex. It is a cyclonic (counter-clockwise on the Northern hemisphere) circumpolar circulation that deepens in fall, when the sun starts to set and temperatures decrease rapidly. The polar vortex separates the cold polar air from warmer mid-latitudinal air. It is weaker in the Northern hemisphere, because the continents and mountain ranges around the Arctic disrupt the circulation. In the stronger Antarctic polar vortex, temperatures fall to extremely low temperatures which causes polar stratospheric clouds to form. The surface of the cloud hydrometeors play a crucial role in destructing stratospheric ozone, leading to the Antarctic ozone hole.

3.2 Arctic amplification

The Arctic is strongly affected by global climate change. Temperatures in the Arctic have increased twice as much compared to global temperatures (see Fig. 3.1), a phenomenon referred to as **Arctic amplification**. Sea ice has retreated significantly in the past decades, see Fig. 3.2 which illustrates the negative trend in minimum sea ice extent and Fig. 3.3 showing the second lowest minimum Arctic sea ice extent observed on September 15th 2020 compared to the median extent of 1981-2010. Several processes are hypothesized to contribute to Arctic amplification (e.g., Serreze and Barry, 2011):

- Albedo feedback: with some initial warming, ice and snow will melt faster and/or earlier than previously. The exposed surface (land or sea) has a lower albedo than ice or snow and absorbs more incoming shortwave radiation. The surface and lower atmosphere is warmed up due to the absorbed incoming shortwave radiation and further melting in the surrounding areas is enhanced, leading to a positive (amplifying) feedback.

- Sea ice loss: sea ice isolates the relatively warm ocean from the cold atmosphere. If the sea ice extent decreases due to initial warming, more heat will be transferred from the ocean to the lower atmosphere, leading to additional warming. This results in a positive feedback loop with even more sea ice melting.

- Horizontal heat fluxes: increased horizontal atmospheric heat fluxes to the Arctic free troposphere have been found to contribute significantly

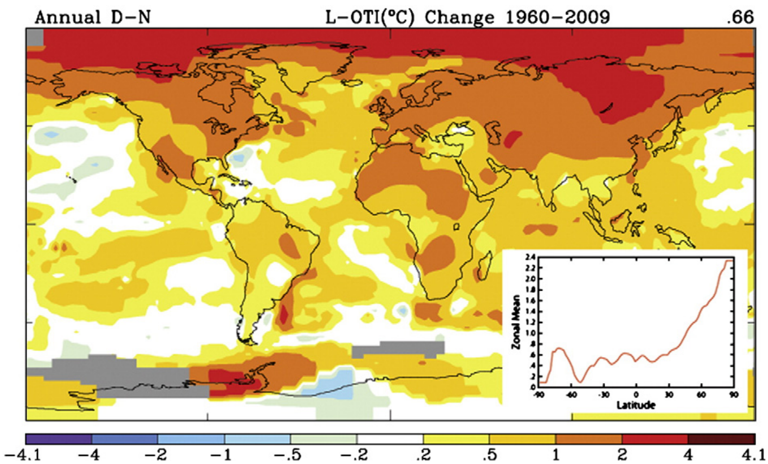

Figure 3.1: Global map of linear trends of temperature change from 1960 to 2009 based on the National Aeronautics and Space Administration Goddard Institute for Space Sciences (NASA GISS) temperature analysis (http://data.giss.nasa.gov/gistemp).
Reprinted from Serreze and Barry (2011), with permission from Elsevier.

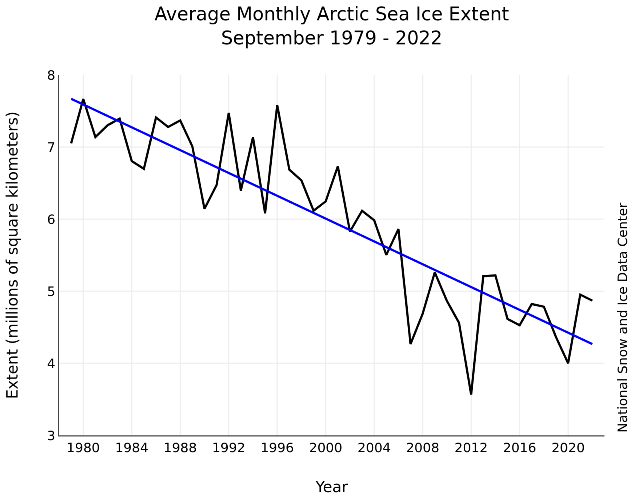

Figure 3.2: Time series of average September Arctic sea ice extent from 1979 to 2022. The linear trend indicated is -12.3% per decade relative to the 1981 to 2010 average.
Image Credit: National Snow and Ice Data Center (2022)

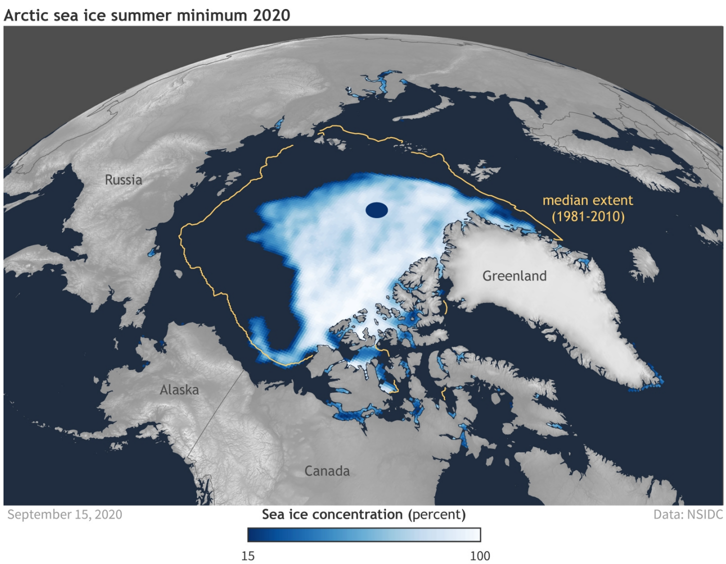

Figure 3.3: Arctic sea ice extent September 15th 2020.
Image Credit: National Oceanic and Atmospheric Administration (NOAA) climate.gov (2020)

to the decadal Arctic warming trend from approximately 1992 to 2008 (Yang et al., 2010). Also in the ocean, heat advection is currently leading to warming in the Arctic (Serreze and Barry, 2011). Both poleward heat flux increases are mainly caused by decadal variations in atmospheric and ocean circulations and thus not caused by anthropogenic emissions. However, they still contribute to the enhanced warming of the Arctic.

- Clouds: Sect. 3.3 describes in more details how clouds impact the Arctic energy balance. In summary, clouds have a warming effect in the Arctic during most of the year. That means if cloud cover would increase, especially for low clouds, there would be a warming effect. Additionally, a change in the clouds' microphysical structure matters - more liquid water in clouds increases the longwave emissions by clouds, leading to a warming at the surface. This increase in liquid water could be caused by the initial warming, basically moving the altitude of cloud ice higher. Observational studies have come to differing conclusions how clouds have changed in different seasons and regions of the Arctic (e.g., Wang and Key, 2005; Eastman and Warren, 2010; Philipp et al., 2020), but overall it appears likely that cloud changes have amplified Arctic warming in the past decades.

- Water vapor: increased water vapor in the Arctic atmosphere leads to enhanced longwave emissions towards the surface and thus warming of the surface. Screen and Simmonds (2010) found that the water vapor content has increased in the Arctic from 1989 to 2008 and that most of this increase can be attributed to sea ice loss which leads to enhanced evaporation from the underlying ocean. Another cause for increased water vapor is the general warming in the Arctic and the consequential increase in saturation vapor pressure.

3.3 Clouds in the Arctic

Clouds have been observed at various Arctic locations during the majority of the year, with a minimum in winter and a maximum in late summer/fall (Shupe et al., 2011). The lower the altitude, the higher their abundance (Shupe et al., 2011).
The specific features of the Arctic climate as described in Sect. 3.1 affect the conditions for clouds. Due to the lower temperatures, ice potentially dominates in clouds. The typically low aerosol concentrations on the other hand limit heterogeneous cloud ice formation between 0 and -38°C. As a consequence, entirely liquid clouds can be observed down to -24°C in the Arctic (Shupe, 2011). However, ice clouds are the dominant cloud phase in the Arctic, followed by mixed-phase clouds (Shupe, 2011). The polar solar cycle and surface properties have large effects on the shortwave Arctic surface energy balance: during winter, the solar part of the energy balance basically disappears. The lower the solar zenith angle, the higher becomes the cooling effect of clouds by reflecting incoming solar radiation (Shupe and Intrieri, 2004). The cooling effect also

increases for decreasing surface albedo, with clouds shielding solar radiation from being absorbed by darker ocean surface. However, these cooling effects lead to a negative net cloud radiative effect only in mid-summer. The positive longwave cloud radiative effect dominates most of the year (Shupe and Intrieri, 2004). The warming greenhouse effect of Arctic clouds is caused by their low altitude (and thus rather high temperature), and is highly sensitive to their water content (Shupe and Intrieri, 2004).
Due to their warming effect, Arctic clouds govern the Arctic surface energy balance, which controls the melting and freezing of sea ice. This means that clouds can have a substantial impact on the cycle of sea ice formation and melting.

3.3.1 Stratiform clouds

Low-level clouds are the most abundant clouds in the Arctic from mid-spring to mid-fall (Curry and Ebert, 1992). Many of the low-level Arctic clouds are stratiform and mixed-phase and they can persist over several hours and up until many days (Shupe et al., 2006). This is a longer lifetime than status clouds typically have in lower latitudes (cf. Sect. 1.1), which means that Arctic mixed-phase stratiform clouds (AMPS) can impact the surface energy balance over an exceptionally long time. Sea ice melting and freeze-up coincide with the highest occurrence of low-level clouds, so AMPS can play a critical role in these transition periods. The vertical structure of AMPS is important for their longevity. Shupe et al. (2008) examined the vertical structure of AMPS and found that liquid water content often reaches its maximum at the cloud top and ice crystals precipitate from this liquid layer. They also found that ice water content increases downwards from the cloud top and peaks near the cloud base, below the cloud most of the ice sublimates before reaching the surface. AMPS mass is dominated by the liquid water with ice water content on average making up only 15% of the mass (Shupe et al., 2008). The dominance of liquid water at the top of the cloud causes cloud-top radiative cooling which is the main driver for the long persistence of AMPS (e.g., Curry, 1986).
Models have difficulties representing AMPS, especially the partitioning between liquid and frozen cloud content (e.g., Morrison et al., 2003; Tjernström et al., 2008; Birch et al., 2012; Stevens et al., 2018). As explained previously however, this partitioning is crucial for modeling the correct evolution and radiative properties of the cloud that can impact the melting and freezing of sea ice.

Chapter 4

Models used to investigate clouds

Researchers use numerical models to investigate the Earth system, project future climate or test different hypotheses related to the Earth system, such as the influence of specific processes on Earth's climate, feedback loops etc. Since clouds are essential for the water cycle and precipitation formation and they have a large impact on the global energy balance, they need to be represented well in models. The same is true for all processes that define the lifetime and characteristics of clouds. We learned in the previous chapters that the phase of the hydrometeors in clouds is an important factor for both precipitation formation and the clouds' radiative properties. This means that the phase of hydrometers, and thus cloud droplet formation and ice nucleation, have to be described realistically in models in order to represent the microphysical structure of the clouds correctly.

4.1 Overview of models

Numerical models that represent the Earth system are based on the conservation of momentum, mass and energy. The equations describing the conservation laws, like the equations of motion, cannot be solved analytically, but they require numerical solutions to calculate the changes with time and space. These numerical methods can differ from model to model and require a discretization of the equations. Another difference between models is the spatial and temporal model resolution. Models exist at a wide range of spatial resolution: General Circulation Models (GCMs) or Earth System Models (ESMs) that are used for climate projections typically have a distance between grid points of 100 km at the equator. Large-eddy simulations (LES) can have grid point-distances as small as a few meters.
Due to the limited spatial resolution, not all processes can be resolved in models and thus have to be parameterized, i.e., the process has to be described based on variables that are resolved on the grid. For example, cloud cover is calculated

based on temperature and relative humidity in the grid boxes. However, which processes have to be parameterized depends on the model used. Turbulence, for example, has to be parameterized in GCMs, while LES models resolve larger turbulent eddies, hence their name. Cloud microphysics, however, always needs to be parameterized, since for example droplet activation or collisions between hydrometeors occur on scales much smaller than the spatial resolution that all models have.

4.2 The LES model MIMICA

The model used in this thesis is the LES model MIMICA (MISU/MIT Cloud-Aerosol model, Savre et al. (2014)). It was developed mainly to simulate high-latitude mixed-phase clouds (e.g., Savre and Ekman, 2015a; Sotiropoulou et al., 2020), but has also been used to investigate mid-latitude marine stratocumulus (e.g., Bulatovic et al. (2019)). In this description of MIMICA, I will focus on the processes described in Chap. 2 and refer to Savre et al. (2014) for a more general description of the model.

4.2.1 Microphysical processes in MIMICA

Cloud microphysics are represented in MIMICA as a two-moment bulk microphysics scheme. 'Two-moment' implies that the hydrometeors are represented by the prognostic variables number concentration, N, and mass mixing ratio, Q, while 'bulk' means that these variables are considered as averaged over the respective hydrometeor type (instead of having size-dependent distributions of N and Q, either continuous or discrete with certain size bins). The mass distributions of all hydrometeors follow regular gamma-distributions. Cloud droplets, rain drops, ice crystals, snow flakes and graupel are the hydrometeors represented in MIMICA.

Latent heat

In the version of MIMICA used here, the latent heats of vaporization, sublimation and melting are constant with temperature. The latent heat of vaporization is given at 25°C, while for sublimation and melting they are valid for 0°C. Latent heat is taken into account by using the ice liquid potential temperature as prognostic variable. For this temperature, basically the latent heat that had been released by the previous condensation of droplets and deposition of ice crystals is subtracted.

Activation of cloud droplets

There are two possibilities to describe the activation of CCN (cloud condensation nuclei, aerosol required for cloud droplet formation) in MIMICA: Aerosol can be represented prognostically, which means that the mass and number densities of different aerosol species are calculated throughout the model run. CCN activation is then treated according to Petters and Kreidenweis (2007) where one parameter describes the hygroscopicity of each aerosol population. For the simulations in Paper 1 we instead chose the option to represent CCN activation according to Khvorostyanov and Curry (2006). Their formulation is based on a power-law depending on the prescribed background CCN concentration and modeled supersaturation.

Ice nucleation

Several options for representing ice nucleation exist in MIMICA. One computationally fast and often used approach is to maintain the ice crystal number concentration N_i within the cloud at a constant specified value. As an example, for Arctic clouds $N_i = 200\ \mathrm{m}^{-3}$ is often used. A number of interactive ice nucleation schemes are also available: an active site scheme (Ickes et al., 2017), the INP concentration parameterization developed by Fletcher (1962), immersion freezing according to Diehl and Wurzler (2004) and heterogeneous ice nucleation following Phillips et al. (2008). The most sophisticated option treats INPs prognostically and follows classical nucleation theory (Savre and Ekman, 2015b). Specifically, I implemented a scheme that draws the INPC from a log-normally shaped relative frequency distribution. If N_i is lower than the picked INPC, new ice crystals are formed (see Paper 1).

Diffusional growth by water vapor, evaporation and sublimation

Diffusional growth by condensation/deposition of water vapor and evaporation/sublimation are modeled following Pruppacher and Klett (1997), based on the combination of diffusion of water vapor to and latent heat of the phase change away from the hydrometeor and vice-versa. The change in Q is proportional to the diameter and number concentration of the respective hydrometeor category and depends additionally on saturation with respect to water/ice and the temperature. As the next step, the evolved supersaturations over water and ice are being calculated, and after that the number of hydrometeors entirely evaporating or sublimating is determined.

Melting

Melting of frozen hydrometeors is calculated according to Pruppacher and Klett (1997) based on temperature and vapor pressure. This representation assumes that first a liquid layer forms on the outer hydrometeor. Melting of an entire hydrometeor results from a balance between the heat transfer through the liquid layer surrounding the frozen center and the supplied latent heat of melting. For the model, the relevant outcome is how many conversions and how much mass change between the different hydrometeor categories occur (e.g., from ice crystal to rain).

Collision processes

Many cold collection processes in MIMICA result in graupel (ice crystal/graupel + cloud droplet, ice crystal/graupel/snow + rain drop, snow + graupel). The

remaining cold collection processes are autoconversion of ice crystals to snow, selfcollection of snow, and growing of snow by colliding cloud droplets or ice crystals. Collisions involving frozen hydrometeors are modeled following Wang and Chang (1993), while collisions of liquid hydrometeors are represented according to Seifert and Beheng (2001) and Seifert and Beheng (2006).
The changes in Q due to warm collision processes are proportional to the mixing ratios of both colliding hydrometeor types, while the changes in N depend on the number concentration of the smaller type and the mixing ratio of the larger type. For cold collection processes, changes in Q and N depend on Q, N, the average fall velocities and diameters of the colliding hydrometeor classes. The terminal fall speed of hydrometeors is calculated from simple power laws of their diameter.

Precipitation

Precipitation fluxes are calculated from fall velocities of rain drops, snow and graupel and their mixing ratios or number concentrations. The surface precipitation is given by the flux in the second vertical level of the model domain.

Secondary ice processes

So far, the only secondary ice process implemented into MIMICA is collisional breakup, where ice crystal fragments result from collisions of different hydrometeors (Sotiropoulou et al. (2021), following Phillips et al. (2017)). Collisions between ice crystals + ice crystal/graupel/snow, snow + graupel/snow and graupel + graupel can lead to additional ice crystal fragments.
The amount and mass of additional ice splinters formed depend on the mixing ratios, number concentrations, average diameters and fall velocities of the colliding hydrometeors.

Chapter 5

Summary of appended papers and outlook

5.1 Paper 1

Heterogeneous ice nucleation parameterizations have three common drawbacks:

- Computational complexity: they might require detailed input from the driving model like aerosol number concentrations, type or size. Whether this input is available depends on the model used, for example which aerosol species are implemented in the model.
- Limited validity: they can be limited to certain conditions of temperature and/or vapor saturation, depending on the measurements they are based on.
- No INPC variability: they often yield one fixed INPC value for a specific set of conditions (temperature, vapor saturation, aerosol concentration, aerosol size and type), which is not representing the full range of observed values.

The parameterization that we propose in this work solves all these problems: It only requires the temperature from the driving model, it is valid for the entire temperature range of heterogeneous freezing and it returns an INPC that is being drawn randomly from a relative frequency distribution (RFD), i.e., it represents the full range and variability of the observations. This INPC RFD is derived from observations that INPC of a given temperature follow a log-normal frequency distribution. The RFD is valid for remote locations.
We tested the new parameterization in a large-eddy simulation of a relatively warm AMPS with in-cloud temperatures between -7 and -10°C using the LES model MIMICA. The ice content of the resulting simulated cloud has a reasonable order of magnitude. Cloud ice content depends linearly on the median INPC of the RFD and exponentially on the standard deviation of the distribution. A test using median INPC instead of random drawing from the

RFD leads to almost no cloud ice and thus highlights the importance of the random drawing. However, cloud ice depends on how often new INPCs are drawn and on model resolution. We thus could show that our parameterization based on the random drawing from an INPC RFD might improve large-eddy simulation of AMPS but still needs amendments (see Outlook).

5.2 Outlook

5.2.1 Continuation on freezing parameterization

The dependence on the frequency of drawing new INPCs and on model resolution have to be further investigated, since both could impede the implementation into other models. Further natural follow-up steps of Paper 1 would be to test the parameterization with different models and for different cloud cases. It is important to test the scheme for cases with lower temperatures since much more ice nucleation is expected to happen. Ideally, for that case reliable observations of cloud ice are available in order to be able to evaluate the parameterization more quantitatively. We would gain more knowledge about the universality of the parameterization by testing it for different cases and in different models. If a different aerosol population is connected with a new test case, the INPC distributions would need to be adapted, since the current formulation assumes maritime aerosol. This could easily be done by modifying the formulation of the INPC RFD (median and width of the distribution). Additionally, it could be tested to define the variability of random drawing in terms of temporal and spatial distance. In the current implementation, a random selection is made for each model grid cell at each time step, and the degree of randomness varies with model resolution.

5.2.2 Studies of other aerosol-induced microphysical aspects in Arctic mixed-phase clouds

The expected further sea ice loss in the Arctic could lead to more shipping traffic in this remote region. Aerosol released by ships can influence cloud formation abilities of Arctic aerosol and thus impact AMPS. We will test the impact of aerosol emitted from different fuel compositions (Santos et al., 2022) on AMPS in large-eddy simulations with MIMICA.

5.2.3 Representation of secondary ice processes in climate models

Apart from heterogeneous ice nucleation and future changes in CCN properties, another big research topic in mixed-phase cloud modeling is secondary ice production (SIP). I will be involved in a project where we will investigate the importance of SIP compared to other processes involving cloud ice in GCMs as part of FORCES - an EU project that aims at improving anthropogenic aerosol radiative forcing for better climate projections (Forces, 2022). We will assess whether the inclusion of SIP in the simulations will lead to ice/liquid apportionment closer to satellite and in-situ observations.

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