Doctoral Thesis

Regional Modeling of Aerosol Processing in Liquid and Mixed-phase Clouds

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Regional Modeling of Aerosol Processing in Liquid and Mixed-phase Clouds

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presented by

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“Le savant n’étudie pas la nature parce que cela est utile;
il l’étudie parce qu’il y prend plaisir
et il y prend plaisir parce qu’elle est belle.
Si la nature n’était pas belle,
elle ne vaudrait pas la peine d’être connue,
la vie ne vaudrait pas la peine d’être vécue.”

“The scientist does not study nature because it is useful to do so.
He studies it because he takes pleasure in it,
and he takes pleasure in it because it is beautiful.
If nature were not beautiful
it would not be worth knowing,
and life would not be worth living.”

Henri Poincaré (1854–1912)
Für meine Eltern.

Pour Sélim.
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Abstract

Atmospheric aerosol particles play an important role for cloud formation as depending on their size, composition and mixing state, they may act as cloud condensation nuclei for the activation of cloud droplets or be effective as ice nuclei for the formation of ice crystals. However, during their residence time in the atmosphere, between their formation or emission and their removal, aerosol particles undergo significant modifications. Physical and chemical processes like coagulation, coating and water uptake and aqueous surface chemistry modify the aerosol composition and size distribution. At this, clouds play a primary role as physical and chemical processing inside cloud hydrometeors contribute considerably to the changes in the aerosol particles. Due to cloud droplet activation, heterogeneous ice nucleation and collision-coalescence processes aerosol particles become incorporated into hydrometeors. Aqueous chemical reactions in liquid droplets can add non- or semi-volatile compounds to the in-hydrometeor aerosol mass. Collision-coalescence processes with other hydrometeors further lead to an accumulation of in-hydrometeor aerosol mass. Eventually, the hydrometeor evaporates / sublimates releasing the collected aerosol material back to the atmosphere. The newly generated aerosol particles are released as larger, internally mixed particles. The described processes are part of the cloud cycling of aerosol particles. Based on theoretical considerations Pruppacher and Jaenicke (1995) calculated that on global average atmospheric particles are cycled three times through a cloud before being removed from the atmosphere. In global simulations of aerosol processing in stratiform clouds only an aerosol particles undergoes on average 0.5 cloud cycles before son removal from the atmosphere (Hoose et al., 2008c).

In order to simulate aerosol processing in clouds on a regional scale, the regional weather forecast and climate model COSMO has been extended by an explicit treatment of in-hydrometeor aerosol particles. The employed model version includes the two-moment cloud microphysical scheme by Seifert and Beheng (2006) which is coupled to the two-moment aerosol module M7 (Vignati et al., 2004) as described in Muhlbauer and Lohmann (2008, 2009) and Zubler et al. (2011a). Five new aerosol modes corresponding to the five hydrometeor classes cloud droplets, ice crystals, rain drop, snow flakes and graupel were implemented in the model and coupled to
the aerosol and the cloud microphysical scheme. In the new modeling framework, only unac-
tivated aerosol particles are attributed to the aerosol module M7, whereas all in-hydrometeor
aerosol particles are treated within the new aerosol modes. The distinction between unacti-
vated and in-hydrometeor aerosol enables the simulation of cloud cycling of aerosol particles. We account for production, transfer and release of in-hydrometeor mass.

The impact of aerosol processing in liquid and mixed-phase orographic clouds on the aerosol population is investigated in an idealized two-dimensional study of moist flow over a double
bell-shaped topography. The results show that aerosol regeneration upon evaporation / sub-
limation replenishes a part of the scavenged aerosol particles. Near to the surface, newly
formed aerosol particles are released in the coarse mode, whereas at higher altitudes primarily
accumulation mode particles are generated. A vertical redistribution of aerosol particles due to
the transport within sedimenting hydrometeors can be observed. The regeneration of aerosol
particles increases the number of cloud droplets upon subsequent cloud formation with pos-
sible implications for the ice crystal number concentration. However, the simulated ice water
content in the mixed-phase cloud is not significantly affected by aerosol processing, as most of
the cloud ice is produced via the Wegener-Bergeron-Findeisen process.

The model performance is demonstrated in two case studies of Arctic liquid and mixed-phase
clouds. Comparison to in-situ observations from the ISDAC flight campaign in Barrow, Alaska
in 2008 allowed to evaluate the new aerosol processing scheme. It is shown that a signifi-
cant number of the scavenged aerosol particles is regenerated by evaporation and sublimation
processes. The regenerated aerosol particles are mostly released to the accumulation mode,
omitting the formation of a significant number of coarse mode particles as seen in the ob-
servations. Sensitivity tests indicate that enhanced contact freezing due to increased number
concentrations of insoluble aerosol particles or the consideration of insoluble bacteria as con-
tact IN allows to better simulate the vertical structure of the ice water content of the observed
cloud.
Zusammenfassung


In idealisierten 2D-Simulationen wurde der Einfluss von Aerosolprozessierung in flüssigphasen und gemischtphasen Wolken auf die Aerosolverteilung untersucht. Die Ergebnisse zeigen,

Chapter 1

Introduction

1.1 Aerosol-cloud interactions

1.1.1 Atmospheric aerosol particles

Aerosol is defined as a dispersion of small solid or liquid particles suspended in air (Seinfeld and Pandis, 2006). This definition comprises particles with large differences in size and composition. Aerosol particles are involved in numerous physical, chemical and microphysical processes and their temporal and spatial distribution in the atmosphere is highly heterogeneous. Aerosol particles can be directly emitted from different sources (primary aerosol) or form via gas-to-particle conversion from precursor gases within the atmosphere (secondary aerosol). The size of aerosol particles can vary over four to five orders of magnitude, from a few nanometers to tens of micrometers. Observations of atmospheric aerosol particles showed that their size spectrum can be described by a superposition of several lognormal size distributions, a concept which takes the size dependent production and removal processes into consideration. Following Whitby (1978), the following modes are commonly distinguished regarding the particle radius:

- nucleation mode \( r < 0.005 \mu m \)
- Aitken mode \( 0.005 < r < 0.05 \mu m \)
- accumulation mode \( 0.05 < r < 0.5 \mu m \)
- coarse mode \( 0.5 \mu m < r \)

Particles with a radius larger than 5 \( \mu m \) are also referred to as giant aerosol particles. The nucleation mode contains the smallest atmospheric aerosol particles with radii up to 5 nm. These fine aerosol particles are formed by gas-to-particle conversion from supersaturated precursor gases, such as the nucleation of sulfate aerosols at low temperature in the upper troposphere.
Due to condensation of vapor and coagulation with other aerosol particles, the fine nucleation mode particles grow into the Aitken mode. Nucleation and Aitken mode particles have the highest number concentrations in the atmosphere, but contribute only little to the total aerosol mass due to their small sizes. Growing further by condensation and coagulation, the particles start to accumulate in the accumulation mode. At accumulation mode sizes, the coagulation process slows down, as neither Brownian motion nor impaction efficiently collide particles. However, the aerosol size is still not large enough for efficient dry deposition by gravitational sedimentation to the surface. Thus, the primary removal processes of accumulation mode particles are activation into cloud droplets and in-cloud and below-cloud scavenging due to collisions with hydrometeors. As a result, accumulation mode particles have a far longer lifetime in the atmosphere than the other aerosol modes. Coarse mode particles mostly originate from primary aerosol sources by bulk-to-particle conversion such as wind blown dust from arid regions, sea salt particles that are formed from evaporating droplets ejected into the air during bubble bursting or sea wave breaking at the ocean surface, large combustion particles such as fly ash and biological particles such as pollen and spores emitted by plants. Due to their increased size, they compose a large fraction of the total aerosol mass. Coarse particles are efficiently removed by dry deposition and wet scavenging processes, and therefore have the shortest lifetime in the atmosphere. Figure 1.1 schematically illustrates the described processes as a function of the aerosol mode.

Another important characteristic of aerosol particles is their mixing state. Due to the different independent aerosol sources, one could think that the aerosol population consists of particles that each contain one pure species which is different from particle to particle (external mixture). However, some particles are already emitted as a mixture of several species (internal mixture), or they become mixed with other compounds upon processing in the atmosphere. In the atmosphere, the mixing state of aerosol particles proves to be complex, consisting of an external mixture of internally mixed particles. Not only the origin, but also the history of the aerosol particles determines the characteristics of the ambient aerosol population such as size, composition and mixing state.

While some aerosol particles originate from natural sources as previously mentioned, other particles are emitted by anthropogenic activities like combustion of fossil fuel. Aerosol particles play an important role in the Earth’s climate system. They impact the radiation budget directly by scattering solar radiation and by absorbing shortwave and longwave radiation. Indirectly, they influence the climate system via their influence on cloud formation, development and properties. Aerosol-cloud interactions represent one of the major uncertainties in the assessment
Some aerosol particles are hydrophobic upon emission, such as BC and partly also POM. Hydrophobic aerosols can only be removed by dry deposition or wet scavenging due to Brownian motion or impaction but not by nucleation scavenging. However, hydrophobic aerosols can acquire soluble material either by colliding with other particles that contain soluble material or if for instance sulfuric acid or organics from the vapor condense onto them. Then they can participate in nucleation scavenging as well.

Figure 1.1: Schematic of the main processes for aerosol particles as a function of their size. AP refers to aerosol particles, BC to black carbon and POM to particulate organic matter.

of climate change (Boucher et al., 2013). The following parts give a more detailed description of the interactions between aerosol particles and clouds and show possible implications for the climate system.

1.1.2 Aerosol effects on clouds

Cloud droplet activation

Homogeneous droplet nucleation of pure water requires far higher supersaturations than observed in natural clouds. In the Earth’s atmosphere, the presence of aerosol particles greatly facilitates the condensation of water vapor allowing for droplet activation at much lower supersaturations.

As many aerosol particles contain some soluble material, small atmospheric water droplets virtually always contain some dissolved solutes such as sodium chloride or ammonium sulfate. To understand the process of the formation of larger cloud droplets, the saturation vapor pressure over such a solution droplet (a water droplet containing some dissolved material) has to be considered. At this, two competing effects play along. First, the saturation vapor pressure over a
pure water droplet is enhanced compared to the one over a flat surface, as the water molecules on a curved surface are more exposed and thus more likely to evaporate. Small droplets with a high surface curvature exhibit a higher saturation vapor pressure than large drops or even flat surfaces. The curvature effect on the saturation vapor pressure is expressed in the Kelvin’s equation. Second, the saturation vapor pressure over a solution is reduced compared to that over pure water. A simple explanation comprises the fact that solute ions cover a part of the droplet surface, shielding that part of the surface from evaporation of water molecules. This effect is described by Raoult’s Law.

To implicitly describe the saturation vapor pressure of an aqueous solution droplet, and thus the conditions under which cloud droplets form, these two competing effects are combined in the Köhler curve (Figure 1.2). Being only accurate for ideal solutions, the Köhler curve illustrates the importance of both effects for different droplet size ranges. For small droplets the solute effect dominates the growth of the droplets, maintaining the droplets in a stable equilibrium. When the droplet size passes the critical radius, the droplets are said to be activated and start to grow spontaneously as the curvature becomes more important. Insoluble, partly wettable aerosol particles may still induce cloud droplet formation by serving as a substrate surface on which the droplet forms.

Many different types of cloud droplet forming aerosol particles, or cloud condensation nuclei (CCN) are present in the atmosphere. Their number concentration is primarily governed by the aerosol number size distribution (Dusek et al., 2006; McFiggans et al., 2006). In clouds, there are usually enough CCN present to keep the supersaturation from rising much above \( \sim 0.1 \) -1 \% (Rogers and Yau, 1989). Under these conditions, only a subset of the aerosol population actually serves as CCN. The droplet activating ability of a single aerosol particle is mainly a function of the number of soluble molecules the particles contains, which in turn depends on the particles size and chemical composition (Andreae and Rosenfeld, 2008). Water-soluble, hygroscopic particles such as sea salt and sulfates, are most suitable for the initiating cloud formation (Pruppacher and Klett, 1997). However, if the supersaturation is high enough, all aerosol particles can activate as cloud droplets. In the size range between 40 and 200 nm, which is most relevant for CCN particles, it was shown that altered assumptions about the organic fraction of the particles can significantly change the CCN number concentration (Quinn et al., 2008). Typical CCN number concentrations range from a few tens particles per \( \text{cm}^3 \) in maritime air to several thousands per \( \text{cm}^3 \) in polluted areas (Seinfeld and Pandis, 2006).
**1.1. Aerosol-cloud interactions**

Cloud droplets do not freeze instantaneously when the temperature falls below 0 °C, but persist as supercooled liquid droplets in the atmosphere until homogeneous freezing occurs at a threshold temperature of around -38 °C (Koop et al., 2000). Statistical fluctuations of temperature and density lead to collisions and aggregation of water molecules eventually forming a stable ice germ (Pruppacher and Klett, 1997) that then induces rapid freezing of the whole droplet. However, a small subset of atmospheric aerosol particles has the ability to serve as ice nuclei (IN) by initiating drop freezing at temperatures well above the onset of homogeneous freezing and/or lowering the required supersaturation. Preferential sites such as phase boundaries or impurities on the particle (active sites) lower the effective surface energy between the new ice phase and the aerosol particle compared to the surface energy between ice phase and parent phase allowing for heterogeneous nucleation to occur at temperatures between -38 °C and 0 °C. The heterogeneous freezing process is particularly important in mixed-phase clouds, where both liquid and frozen water coexist. These clouds are frequent in mid-latitude...
and polar regions and play an important role in the hydrological cycle since most of the precipitation forms via the ice phase (Lau and Wu, 2003). The air in mixed-phase clouds is saturated with respect to supercooled liquid droplets and thus supersaturated with respect to ice. Due to the difference in saturation vapor pressure over liquid and ice, existing ice crystals can rapidly grow at the expense of supercooled droplets, which evaporate once the relative humidity drops below water saturation. This process, which is called Wegener-Bergeron-Findeisen (WBF) process (Wegener, 1911; Bergeron, 1935; Findeisen, 1938), allows precipitation formation to be more efficient in mixed-phase than in warm clouds. However, to initiate the WBF process some ice crystals have to be formed first. Following Vali (1985), four different mechanisms of heterogeneous freezing, namely, deposition nucleation and immersion, condensation and contact freezing are commonly distinguished. The processes are schematically illustrated in figure 1.3. As the activation of liquid cloud droplets occurs on CCN in the atmosphere (section 1.1.2), there

![Figure 1.3: Schematic of the different freezing modes as a function of ice saturation ratio $S_i$ and temperature $T$ (Hoose and M"ohler, 2012). The solid line indicates saturation with respect to water, the dashed line homogeneous freezing of solution droplets according to Koop et al. (2000).](image)

is always some foreign material inside the droplet. Solid, insoluble particles immersed in the drop have the potential to trigger the phase transition from water to ice during the cooling process if temperatures are sufficiently low. Due to immersion of the ice nuclei in the preexisting droplet at warmer temperatures, this mechanism is called immersion freezing. Condensation
1.1. Aerosol-cloud interactions

freezing also requires relative humidities above water saturation at supercooled temperatures. It refers to the temporarily formation of a water phase by condensation of water vapor on the IN and instantaneous freezing without further cooling. Therefore, the involved aerosol particles have to exhibit properties of an efficient CCN and IN at the same time. Under atmospheric conditions, the differentiation between immersion and condensation freezing remains challenging. Collisions between IN and supercooled droplets can initiate droplet freezing in the contact freezing mode by reducing the energy barrier for ice nucleation. It has also been proposed that "inside-out" contact nucleation may take place upon evaporation, as the shirking droplet surface may eventually get in contact with an immersed IN (Durant and Shaw, 2005). Laboratory studies indicate that contact freezing has higher onset temperatures than the other freezing modes (Ladino et al., 2013). Deposition nucleation refers to the adsorption of water vapor onto the IN surface followed by an immediate phase transition into ice. Therefore, it only requires supersaturation with respect to ice, allowing for a nucleation below water saturation. However, the mechanism is less efficient in mixed-phase clouds, where liquid and ice coexist, as for given supersaturated conditions, the critical radius of an ice embryo in liquid water is smaller than in vapor (Cooper, 1974) favoring immersion freezing compared to deposition nucleation. The onset temperatures of the different freezing modes depend on the nature of the initiating ice nuclei.

Not all solid particles enhance ice formation, but several materials have shown to be ice active (Cziczo et al., 2009b). Ice-nucleating particles are often identified as solid, water insoluble particles (Pruppacher and Klett, 1997) such as fly ash, mineral dust or soot. These properties are different from the characteristics which are associated with good CCN. The freezing onset temperatures depend on the freezing mode as well as the involved IN particle. The actual characteristics which determine the ice nucleation ability of a specific material are still not well known. For a single particle, its surface structure including its size, active sites and ability to form hydrogen bonds are most important to determine its ability to act as IN (Pruppacher and Klett, 1997). It has been hypothesized that a structure similar to the crystal lattice of ice may favor ice nucleation. Many experimental studies focused on the investigation of the ice-nucleating ability of different aerosol types (e.g. Fornea et al., 2009). The same particle may nucleate ice by different freezing modes, which makes the exact characterization of IN difficult. In a review, Hoose and Möhler (2012) give an overview over IN active particles. Mineral dust particles are reported to be efficient ice nuclei in the atmosphere. Soot particles are also investigated in several studies. They reveal to be less efficient ice nuclei than dust particles, as they require higher ice-supersaturations for deposition nucleation and lower temperatures
for immersion freezing. Other candidates for ice initiation represent biological particles such as pollen and bacteria. A small part of bacterial strains and fungal species have been shown to possess an ice nucleation active protein, which allows them to trigger ice formation at high subzero temperatures. There are reports of ice nucleation due to other species like some organic acids, and occasionally of volcanic fly ash and sea salt. Metallic particles were also found to have ice nucleating ability (Cziczo et al., 2009b) and were observed in atmospheric ice residuals (Cziczo et al., 2013).

Aging and cloud processing can change the ice nucleation activity of an aerosol particle. Soluble material condensed on the surface of an IN (e.g. coating of mineral dust with sulfate) can make the IN unavailable for deposition nucleation or contact freezing where a dry aerosol particle is required (Cziczo et al., 2009a). In contrast, Kanji et al. (2013) reported an enhancement of the ice nucleation ability of mineral dust due to ozone aging for low effective aging times. However, the response to ozone aging is not linear, as the ice nucleation ability for high effective aging times was reduced.

1.1.3 Cloud effects on aerosols

During their residence time in the atmosphere, between their emission and their removal, aerosol particles undergo significant modifications. Processes like coagulation of aerosol particles, coating by condensation of gaseous species on the aerosol particle or water uptake alter the aerosol size distribution and composition. At this, clouds play a primary role, as physical and chemical processing within clouds contributes considerably to the change in the aerosol population.

By acting as nucleation centers for droplet or ice formation (section 1.1.2), aerosol particles are scavenged from the atmosphere and incorporated into cloud droplets or ice crystals (activation scavenging). Collisions between aerosol particles and the various hydrometeors further lead to an accumulation of aerosol mass inside the hydrometeors (Pruppacher and Klett, 1997). These collisions can take place as in-cloud impaction scavenging within the cloud, or as below-cloud scavenging by sedimenting hydrometeors which pick up aerosol particles when they fall. In-cloud and below-cloud collision scavenging, covered by the term wet deposition, remove 80-90 % of the mass of fine aerosol particles (Textor et al., 2007).

Within liquid droplets, aqueous chemical reactions can take place. The soluble part of the aerosol mass dissolves in the water. Atmospheric precursor gases may enter the liquid phase by diffusion and subsequently dissociate into ions allowing for aqueous chemical reactions.
1.1. Aerosol-cloud interactions

together with the dissolved aerosol mass. Upon evaporation, dissolved material becomes part of the aerosol phase. A prominent example for aqueous chemistry in droplets is the oxidation of sulfur dioxide to form sulfuric acid. Due to collision-coalescence between two hydrometeors aerosol mass is accumulated in the remaining hydrometeor.

When precipitating hydrometeors eventually reach the ground, the collected aerosol mass is removed from the atmosphere. However, a large fraction of the droplets and crystals evaporates / sublimes releasing the incorporated aerosol mass back to the atmosphere. Laboratory studies of evaporating droplets showed that upon drop evaporation only one single aerosol particle remains (Mitra et al., 1992). Due to dissolution and adherence, the dissolved soluble material concentrates in the liquid phase or crystallizes upon evaporation, so that the released aerosol particles is composed of an internal mixture of all soluble and insoluble aerosol mass within the droplet (Pruppacher and Klett, 1997). Following this, the new aerosol particles has an increased mass and possibly altered composition and mixing state compared the original particle before cloud processing. A schematic of the in-hydrometeor aerosol particles and the most important processes is given in figure 1.4. The schematic considers five different hydrometeor types which are cloud droplets, ice crystals, rain, snow and graupel.

Figure 1.4: Schematic of the different in hydrometeor aerosol particles including microphysical source, transfer and removal processes.
Changes in the aerosol properties due to cloud processing become apparent in substantial modifications of the aerosol size distribution. Observations of aerosol processing in clouds translate in observations of modified aerosol size distributions or altered aerosol composition. Hoppel et al. (1986) and Hoppel et al. (1990) measured bimodal aerosol size distributions in the marine boundary layer which they attributed to cycling in non-precipitating clouds. Considering the history of the air masses, they concluded that the larger mode is formed by activated and cloud processed aerosol particles, whereas the fine mode constitutes of freshly nucleated particles. Air masses which did not pass cloudy regions did not show this bimodal signature.

A distinct bimodal shape of the aerosol size distribution, probably induced and maintained by processing of aerosol particles within clouds, was also observed at the high Alpine research station Jungfraujoch in the Swiss Alps (Weingartner et al., 1999). On the downstream side of a hill cap clouds, Bower et al. (1997) observed a significant modification of the aerosol size distribution and hygroscopic properties due to cloud processing within the cap cloud.

Eck et al. (2012) studied size distribution retrievals after cloud dissipation from the Aerosol Robotic Network (AERONET). Some of the retrievals of cloud processed aerosol particles revealed to be bimodal in the accumulation mode, where the larger mode could be associated with cloud processed aerosol particles and the smaller mode attributed to interstitial aerosol particles which were not subject to modification by incorporation in droplets. Investigating Caribbean trade cumulus clouds, Rauber et al. (2013) found that vertical transport and cloud processing, independently from any humidification effect, contributed to the modifications of the aerosol size distribution within the cloud margins, particularly close to the cloud boundaries. At this, chemical reactions in the aqueous phase may play an important role, as they transfer mass from precursor gases to the aerosol phase. The solubility of the precursor gases in the cloud droplets may be considerably enhanced by the influence of aqueous reactions as demonstrated by Zhang and Tie (2011) based on in-situ measurements of sulfur dioxide. Li et al. (2011) collected in-situ observations of aerosol size distributions and compositions under highly polluted aerosol conditions in China. Cloud residues were found to be significantly larger than interstitial aerosol particles and their composition was dominated by sulfates and soluble organic matter. However, these measurements do not allow to differentiate between the effect of particle selection upon droplet nucleation and the impact of cloud processing on the population of cloud residuals.

In a theoretical approach, Pruppacher and Jaenicke (1995) estimated that an aerosol particle sampled at a distant location from its source region has been cycled three times through stratiform or convective clouds before being removed from the atmosphere. Global simulation of
1.1. Aerosol-cloud interactions

cloud cycling though stratiform clouds only resulted in 0.5 cloud cycles for an average aerosol particles (Hoose et al., 2008b), which the authors claimed to be in agreement with a recalculation of Pruppacher and Jaenicke’s (1995) approach for stratiform clouds only.

1.1.4 Modeling of aerosol processing in clouds

To account for aerosol aging in the atmosphere, many models explicitly account for coagulation of aerosol particles, reducing the number concentration and augmenting the size of the particles during their residence time in the atmosphere. When differentiating between internally mixed and insoluble aerosol particles, such as freshly emitted dust and black carbon, an aerosol scheme may also simulate atmospheric aging like coating with sulfate by transferring insoluble particles into a soluble, mixed mode. Some models also account for gas-to-particle conversion of sulfur dioxide to form sulfate aerosol particles in the upper troposphere.

When it comes to the modeling of aerosol processing within clouds, the complexity of the considered processes varies substantially from one model to another. Aqueous chemical reactions in cloud droplets may involve numerous dissolved precursor gases which substantially increase the complexity of the model. In simple approaches, only the formation of sulfate by oxidation of sulfur dioxide in cloud droplets is parameterized. The created sulfate mass may be added to the gas phase sulfate mass and then partitioned to the aerosol phase via equilibrium, or may be directly attributed to the aerosol particles (e.g. Gong et al., 2011). Sulfate containing droplets may change the properties of insoluble mineral dust aerosol particles significantly. Simulations of multiple cloud cycles of an air parcel model showed that due to impaction scavenging of insoluble dust particles with sulfate containing droplets and subsequent droplet evaporation, larger sulfate coated dust particles are generated, which may serve as giant cloud condensation nuclei (GCCN) for further cloud formation (Wurzler et al., 2000). Appearing as a tail of the size distribution for larger sizes, these particles also lead to a shift of the droplet spectrum towards larger sizes. The more cloud cycles an aerosol particles undergoes, the more its size increases due to collisions with other droplets, impaction scavenging of other aerosol particles and scavenging of gases.

In studies of aerosol processing in convective clouds it was found that aqueous production of sulfate contributes only little to the total hydrometeor aerosol mass. However, processed aerosol particles have been shown to significantly affect the CCN number concentration, delaying drop freezing or reducing crystal growth (Yin et al., 2005) with implications for precipitation formation.
Mechem et al. (2006) considered warm-phase cloud processing of CCN particles including drop nucleation, autoconversion and accretion as collision-coalescence processes of cloud droplets and regeneration of CCN by evaporating droplets in a regional model. However, the CCN population in the model is represented by a single number concentration.

Instead of adding the regenerated aerosol particles to the CCN number concentration, Ivanova and Leighton (2008a) introduced two new aerosol modes in their model, representing regenerated in-cloud droplet and regenerated in-rain aerosol particles. As the particles of these modes can serve as CCN and become activated, cloud cycling becomes possible. Cloud processing like solute transfer due to collision-coalescence between hydrometeors, aqueous chemistry and the release of aerosol particles upon evaporation can either enhance or suppress the number of activated droplets in further cloud cycles (Ivanova and Leighton, 2008b).

Recently Xue et al. (2010) extended their aerosol bin-microphysical module by 40 more aerosol bins to account for increased sizes of aerosol particles due to cloud processing. Based on the number of regenerated aerosol particles and the subsequent reconstruction of the corresponding aerosol size distribution, the regenerated aerosol particles are reattributed to the aerosol module where they can initiate droplet formation in a further cloud cycle. As a result, the number concentration of droplets increased due to the additional CCN particles. In warm-phase clouds, the rain formation was suppressed due to decreased collision-coalescence rates of the smaller droplets.

The described parameterizations only include cloud cycling of aerosol particles acting as CCN. However, in the atmosphere, the ice phase also plays an important role. Xue et al. (2012) investigated the effect of recycled CCN on mixed-phase clouds, where they influenced the riming efficiencies with implications for precipitation formation. However, ice nucleation also plays an important role in mixed-phase clouds. At this, the cycling of IN may change ice nucleation ability of aerosol particles substantially. Therefore, it is important to consider cloud processing not only of CCN, but of all aerosol particles with implication for CCN and IN concentrations. For this, not only the parametrization of cloud droplet formation, but also the freezing parameterizations have to depend on the processed aerosol population.

Hoose et al. (2008a) elaborated an explicit and detailed treatment for cloud droplet and ice-crystal borne aerosol mass for the global circulation model ECHAM5-HAM. This scheme allows to consider cloud processing of aerosol particles in the liquid and the ice phase. The distinction between liquid and ice phase allows to account for mixed-phase processes such as the WBF process. Recycled aerosol particles are reattributed to the unactivated aerosol modes, where
they can serve anew as IN or CCN for further cloud formation.

### 1.2 Aerosols in the climate systems

Aerosol particles influence the Earth's climate by impacting the radiation budget and hydrological cycle. In general, aerosol concentrations are lower over the ocean than over land, ranging from about $20 \text{ cm}^{-3}$ under clean polar conditions to more than $100000 \text{ cm}^{-3}$ in urban pollution (Seinfeld and Pandis, 2006). Human activities change the Earth's climate not only by emission of greenhouse gases, but also via emissions of aerosol particles and their precursor gases. As a result, the aerosol number concentration is increased and the aerosol composition may be modified. Aerosol particles directly impact the Earth's energy budget by interaction with radiation (radiative forcing due to aerosol-radiation interactions). Due to their ability to initiate droplet and ice formation, they affect the radiative balance indirectly by affecting the formation and properties of clouds (Lohmann and Feichter, 2005). These indirect aerosol effects (radiative forcing due to aerosol-cloud interactions) represent one of the largest uncertainties in the climate system (Boucher et al., 2013).

#### 1.2.1 Radiative forcing due to aerosol-radiation interactions

Aerosol particles directly influence the Earth's radiation budget by scattering and absorbing radiation. At this, they impact incoming solar radiation as well as the outgoing thermal radiation. Their impact strongly depends on the aerosol optical properties as well as on the location, the environmental conditions and the underlying surface. The "semi-direct" effect considers the absorption of incoming shortwave radiation by tropospheric aerosol leading to a heating of the troposphere with implications for the relative humidity profile and the stability of the troposphere. This could inhibit cloud formation and / or prompt existing droplets to evaporate.

#### 1.2.2 Radiative forcing due to aerosol-cloud interactions

Due to their droplet and ice nucleating ability, aerosol particles may influence the presence, abundance, lifetime and properties of clouds. In this way, they influence the radiative balance indirectly via their impact on clouds. Depending on the phase of the cloud (liquid, mixed-phase, or ice cloud), different mechanisms, also known as indirect aerosol effects, can be active (figure 1.5).
Cloud albedo effect

Increased aerosol number concentrations may increase the CCN number concentration. At a fixed liquid water content, a greater number of CCN lead to enhanced droplet number concentration, but reduced mean droplet size (Twomey et al., 1974). This then increases the cloud albedo, as more incoming solar radiation is reflected back to space.

Cloud lifetime effect

Increased aerosol concentrations result in a greater number of smaller cloud droplets which have a reduced collision efficiency, and thus reduced collision-coalescence rates, slowing down the formation of rain. Decreased precipitation formation rates translate in a prolongation of the cloud lifetime and hence an increase in the cloud reflectivity over time (Albrecht, 1989). Drizzle suppression together with increased cloud droplet concentrations and decreased droplet radii were observed in marine stratus regions polluted by aerosol emissions from ships (Ferek et al., 2000; Lu et al., 2009). However, based on large eddy simulations and in-situ observations
of shallow clouds Jiang et al. (2006) and Small et al. (2009) find that the cloud lifetime is not significantly increased by augmented aerosol concentrations. They assume that the lifetime of a polluted cloud can be reduced by enhanced evaporation of smaller droplets.

**Riming effect**

The existence of more, but smaller droplets also has an important effect on mixed-phase clouds. After initially growing by diffusion of water vapor deposition, atmospheric ice particles grow by collision with supercooled droplets. During this riming process sedimenting ice hydrometeors collect supercooled droplets which freeze onto the crystals surface. The collision efficiency between ice hydrometeors and droplets decreases for smaller droplets reducing the riming rate (Pruppacher and Klett, 1997). While a reduction of both the riming and snowfall rate due to increased anthropogenic aerosol concentrations were observed in orographic clouds over the Rocky Mountains (Borys et al., 2003), global simulations could not confirm a decrease of the snowfall rate on a global scale (Lohmann, 2004).

**Glaciation effect**

A small part of aerosol particles can serve as nuclei for ice formation. Anthropogenic emissions (e.g. soot emissions) may increase the number of ice-nucleating particles in the atmosphere leading to an increase in the glaciation of supercooled mixed-phase clouds and and enhancement of precipitation formation via the ice phase (Lohmann, 2002a). This effect may partly counteract the effect of increased aerosol number concentrations in warm clouds. However, given newer studies the nucleating ability of soot is controversial (Hoose and Möhler, 2012).

**Deactivation effect**

On the other hand, natural ice nuclei may be deactivated by anthropogenic gaseous emissions such as sulfur dioxide, nitrogen dioxide or ammoniac. For a given ice nucleating material, immersion freezing generally occurs at lower temperatures than contact freezing. Due to coating with hygroscopic material such as anthropogenic sulfuric acid, an insoluble, highly active contact IN may be inhibited to act in the contact mode. The coated particles can be converted into immersion mode nuclei, which would still initiate freezing but at lower temperatures (Hoose et al., 2008c; Storelvmo et al., 2008). Laboratory experiments have shown that in the deposition mode, soot coated with sulfate nucleates ice less efficiently than uncoated soot (Möhler et al., 2005). Recent studies showed that sulfuric acid coatings reduced the ice nucleation abilities of different mineral dusts (e.g. Cziczo et al., 2009a; Chernoff and Bertram, 2010).
1.3 Motivation

The International Panel on Climate Change (IPCC) has stated that the climate is changing and anthropogenic emissions are responsible for most of the climate change over the last 50 years. To accurately assess human-induced climate change, the different components of the climate systems and their interactions have to be understood. At this, aerosol-cloud interactions still represent one of the largest uncertainties in the climate system (Boucher et al., 2013).

Many scientific studies concentrated on the aerosol-cloud interactions in warm-phase clouds. The enhancing effect of increased aerosol concentrations on cloud albedo, counteracting global warming due to greenhouse gas emissions, is consistent in most findings. However, due to their prevalence and importance for precipitation formation, mixed-phase clouds are just as fundamental in the climate system. The interactions of aerosol particles with these clouds are far more complex. The impact of anthropogenic emissions on IN number concentrations and thus on heterogeneous ice nucleation processes remains poorly understood. To investigate these effects, it is crucial to consider aerosol processing within clouds, as it may change the aerosol properties and thus the number concentration of available CCN and IN with implications for further cloud formations and cloud properties.

The aim of this PhD work is to refine the representation of aerosol particles in the regional weather forecast and climate model COSMO by implementing an explicit scheme for cloud-borne aerosol particles. This new parameterization includes a detailed treatment of cloud microphysical processes and their implications for in-hydrometeor aerosol particles and allows for the simulation of cloud cycling of aerosol particles. By means of this new scheme, we analyze the impact of microphysical processing in liquid and mixed-phase orographic clouds on the aerosol size distribution in an idealized setup. Thereafter, we investigate the impact of aerosol processing in Arctic liquid and mixed-phase clouds.
Chapter 2

Model description

2.1 The regional climate model COSMO-CLM

The COSMO model is a non-hydrostatic, fully compressible, limited-area atmospheric prediction model. Its applications comprise both operational numerical weather prediction and various scientific applications on the meso-scale including future climate simulations. The model, originally developed by the German weather service (DWD), is currently developed and applied within the COnsortium for Small-scale MOdeling (COSMO, http://www.cosmo-model.org). The simulations in the present study are performed using the climate version of the model, COSMO-CLM, also known as CCLM, and formerly CLM, release 4.0 (http://www.clm-community.eu).

The model is based on the primitive hydro-thermodynamical equations which are formulated on a rotated Arakawa C-grid (Doms and Schättler, 2002; Steppeler et al., 2003). A split-explicit third order Runge-Kutta scheme is used for time integration in combination with a 5th order upstream horizontal advection scheme. The aerosol and moisture variables are advected by the second order Bott scheme (Bott, 1989).

The employed model version includes a two moment bulk cloud microphysics scheme (Seifert and Beheng, 2006) which is coupled to the two-moment aerosol module M7 (Vignati et al., 2004). The aerosol module M7 has also been used within the framework of the ECHAM-HAM GCM (Stier et al., 2005). The following subsections provide more detailed information on the treatment of the cloud and aerosol microphysics in the model.
2.2 Cloud microphysics

Cloud microphysical processes are treated within the two-moment bulk scheme for water and ice clouds from Seifert and Beheng (2006). The scheme includes prognostic equations for mass and number densities of five different hydrometeor classes: cloud droplets, ice crystals, rain drops, snow flakes and graupel. The number density size distributions $N_{\text{hydro}}(m)$ are represented by generalized gamma-distributions of the form

$$N_{\text{hydro}}(m) = A m^{\nu_{\text{hydro}}} \exp(-\lambda \mu_{\text{hydro}})$$

(2.1)

Here, $m$ denotes the hydrometeor mass, and the index $\text{hydro} \in \{c, r, i, s, g\}$ denotes the hydrometeor class, which can be either cloud droplets ($\text{hydro} = c$), rain drops ($\text{hydro} = r$), cloud ice ($\text{hydro} = i$), snow ($\text{hydro} = s$) or graupel ($\text{hydro} = g$). The two free parameters $\nu_{\text{hydro}}$ and $\mu_{\text{hydro}}$ are kept fixed for each hydrometeor class. The coefficients $A$ and $\lambda$ are derived from the number and mass densities as described in Seifert and Beheng (2006).

2.2.1 Warm-phase processes

Warm-phase cloud processes comprise all cloud microphysical processes involving exclusively liquid hydrometeors. These processes not only contribute directly to surface precipitation, but more importantly, they represent a prerequisite for subsequent ice-phase processes which are thought to be a major catalyst for precipitation formation in the mid-latitudes.

The cloud microphysics scheme of Seifert and Beheng (2006) distinguishes between cloud droplets and rain drops by decomposing the liquid drop distribution into two portions. The threshold is a given drop mass of $2.6 \cdot 10^{-10} \text{kg}$ which corresponds to a radius of $40 \, \mu\text{m}$.

The scheme accounts for activation of cloud droplets, condensational growth and evaporation of cloud droplets, autoconversion (formation of rain by coagulating cloud droplets) and accretion (growth of rain drops by collecting cloud droplets), selfcollection (mutual coagulation of cloud droplets / rain drops, remaining in the same drop category), evaporation of rain and collisional break-up of large rain drops as discussed by Seifert and Beheng (2006).

Sedimentation of rain is parameterized using mass and number weighted mean fall velocities. Due to the lower air density in higher altitudes, the terminal fall velocity is increasing with height. For rain drops the terminal fall velocity is approximated by an empirical relation similar to Rogers et al. (1993), whereas the sedimentation of cloud droplets is based on the Stokes’ terminal fall velocity.
2.2 Cloud microphysics

2.2.2 Ice-phase processes

Ice-phase processes comprise all microphysical processes involving at least one ice-phase hydrometeor. The present cloud microphysics scheme differentiates between cloud ice, snow and graupel particles. The diameter-mass as well as velocity-mass relations of the different particles are parameterized by power laws of the form

\[ D_h(m) = a m^b \]  
\[ v(m) = \alpha m^\frac{\beta}{\gamma} \left( \frac{\rho_0}{\rho} \right)^\gamma \]

where \( \rho \) denotes the air density and \( \rho_0 = 1.225 \text{ kg m}^{-3} \). The coefficients \( a, b, \alpha, \beta, \gamma \) are specified for each hydrometeor class in Seifert and Beheng (2006). Cloud ice is considered as "hexagonal plates" (Heymsfield and Kajikawa, 1987), snowflakes are assumed to be "mixed aggregates" (Locatelli and Hobbs, 1974) and for graupel, a "lump graupel" geometry is used (Heymsfield and Kajikawa, 1987). The size distributions are represented by generalized gamma-distributions (equation 2.1) based on the corresponding shape parameters.

Microphysical processes including the ice-phase are homogeneous and heterogeneous freezing, diffusional growth of ice crystals, aggregation (formation of snow by coagulating ice crystals), self-collection (mutual coagulation of ice / snow / graupel, remaining in the same hydrometeor category), riming (coagulation between the liquid and the different ice phases), melting and sublimation. Related secondary processes e.g. Hallet-Mossop ice multiplication, partial conversion of riming cloud ice to graupel, riming splintering and enhanced melting are accounted for as well. The Wegener-Begeron-Findeisen process is represented implicitly in the model, as in an air parcel, which is supersaturated with respect to ice, but subsaturated with respect to water, all condensate evaporates and the ice crystals grow by water diffusion.

Sedimentation of cloud ice, snow and graupel follows the same approach as for cloud droplets and rain drops. The mean fall velocities can be derived from the velocity-mass relationships and the size distributions. We refer to Seifert and Beheng (2006) for a detailed description of the different cloud microphysical processes.
2.3 Aerosol microphysics

The cloud microphysics scheme has been coupled to the M7 aerosol module (Vignati et al., 2004; Muhlbauer and Lohmann, 2008) which is also used within the framework of the general circulation model system ECHAM5-HAM (Stier et al., 2005).

The M7 aerosol module includes prognostic equations for the number density and mass densities of seven different aerosol classes. The module accounts for four internally mixed, soluble particles (nucleation, Aitken, accumulation, and coarse mode), containing both soluble and insoluble compounds, and three insoluble modes (Aitken, accumulation, and coarse mode), which are characterized by low water solubility. The aerosol module considers five different aerosol compounds, including sulfate (SU), carbonaceous aerosols (black carbon BC, organic carbon OC), sea salt (SS) and mineral dust (DU). At this, the insoluble modes contain externally mixed freshly formed black carbon and dust. Due to sulfate coating or coagulation, insoluble aerosols can be transferred to the mixed modes.

The modal concept describes the aerosol population by a superposition of seven lognormal size distributions of the form

$$N(\ln r) = \frac{\sum_{k=1}^{7} N_k}{\sqrt{2\pi} \ln \sigma_k} \exp \left[ -\left( \frac{\ln r - \ln \bar{r}_k}{\sqrt{2 \ln \sigma_k}} \right)^2 \right]$$

(2.4)

Here, $N_k$ denotes the aerosol number concentration of the $k$-th aerosol mode with $k \in [1, 7]$. $\sigma_k$ denotes the geometric standard deviation representing the width of the particle size distribution of the $k$-th mode. For the standard deviation we adopt fixed values based on Wilson et al. (2001): $\sigma_k = 1.59$ for the nucleation, Aitken and accumulation mode and $\sigma_k = 2.0$ for the coarse mode. Varying within the given size range (table 2.1), the count median radius $\bar{r}_k$ of the $k$-th mode is determined from the number and mass densities following

$$\bar{r}_k = \sqrt[3]{\frac{3}{4\pi N_k} \sum_{\text{subst}} M_k^{\text{subst}} \frac{\rho_{\text{subst}}}{\rho_{\text{subst}}} \exp \left( -\frac{3 \ln^2 \sigma_k}{2} \right)}$$

(2.5)

for all aerosol modes $k \in [1, 7]$, aerosol compounds $\text{subst} \in \{\text{SU, BC, OC, SS, DU}\}$ and aerosol densities $\rho_{\text{subst}}$.

The aerosol module M7 accounts for nucleation of gas-phase sulfuric acid (Vehkamäki et al., 2002), condensation of sulfuric acid on pre-existing aerosol particles, coating of insoluble aerosol particles with sulfuric acid, inter- and intramodal coagulation and water vapor uptake.
### 2.3. Aerosol microphysics

<table>
<thead>
<tr>
<th>Aerosol mode</th>
<th>Size range [µm]</th>
<th>Number mode</th>
<th>Mass modes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nucleation mixed mode</td>
<td>&lt; 0.005</td>
<td>$N_1$</td>
<td>$M_{SU}^{1}$</td>
</tr>
<tr>
<td>Aitken mixed mode</td>
<td>0.005 – 0.05</td>
<td>$N_2$</td>
<td>$M_{SU}^{2}$, $M_{BC}^{2}$, $M_{OC}^{2}$</td>
</tr>
<tr>
<td>Accumulation mixed mode</td>
<td>0.05 – 0.5</td>
<td>$N_3$</td>
<td>$M_{SU}^{3}$, $M_{BC}^{3}$, $M_{OC}^{3}$, $M_{SS}^{3}$, $M_{DU}^{3}$</td>
</tr>
<tr>
<td>Coarse mixed mode</td>
<td>&gt; 0.5</td>
<td>$N_4$</td>
<td>$M_{SU}^{4}$, $M_{BC}^{4}$, $M_{OC}^{4}$, $M_{SS}^{4}$, $M_{DU}^{4}$</td>
</tr>
<tr>
<td>Aitken insoluble mode</td>
<td>0.005 – 0.05</td>
<td>$N_5$</td>
<td>$M_{OC}^{5}$, $M_{DU}^{5}$</td>
</tr>
<tr>
<td>Accumulation insoluble mode</td>
<td>0.05 – 0.5</td>
<td>$N_6$</td>
<td>$M_{DU}^{6}$</td>
</tr>
<tr>
<td>Coarse insoluble mode</td>
<td>&gt; 0.5</td>
<td>$N_7$</td>
<td>$M_{DU}^{7}$</td>
</tr>
</tbody>
</table>

Table 2.1: Table of the different aerosol modes of the aerosol module M7. (The size range of each mode is given for the count median radius $\bar{r}_k$. $N_k$ and $M_k^{\text{subst}}$ correspond to the aerosol number and mass density for the mode $k \in [1, 7]$ and compound $\text{subst} \in \{SU, BC, OC, SS, DU\}$ respectively.)

For a detailed description of the parameterized processes we refer to Vignati et al. (2004), Stier et al. (2005) and Muhlbauer and Lohmann (2008).

In the standard version of the model, the M7 aerosol module comprises the total aerosol population, including both, free unactivated particles and activated in-hydrometeor aerosols. The here presented new model version assigns only the unactivated aerosol particles to the M7 module. For hydrometeor-borne aerosol mass five new modes corresponding to the five prognostic hydrometeor classes were introduced into the model following the approach of Hoose et al. (2008a). We distinguish between in-cloud droplet (CD), in-ice crystal (IC), in-rain drop (RD), in-snow flake (SF) and in-graupel (GR) aerosol mass. The five new modes can contain a contribution of all five aerosol components (SU, BC, OC, SS, DU) (Table 2.2). The number densities of hydrometeor-borne aerosol particles are not prognostic variables, but are diagnosed from the corresponding hydrometeor number densities. All material which is taken up by a hydrometeor is transferred to the new modes CD, IC, RD, SF and GR. The new modes are not limited by any specific size range. Their median radii are calculated as described below (equation 2.23). In the standard model version, aerosol and cloud microphysics are coupled in a way that the aerosol number concentrations influence the cloud properties, but are not changed themselves. The new scheme tracks aerosol particles even when incorporated into hydrometeors allowing for the simulation of cloud cycling of aerosol particles. This enables to simulate interaction between aerosols and clouds in a complete way.

Aerosol particles are incorporated into hydrometeors due to the different scavenging processes, including activation of aerosol particles, in-cloud impaction scavenging and below-cloud scavenging by precipitating hydrometeors. Once inside a hydrometeor, the aerosol particles un-
Aerosol mode | Abbreviation | Mass modes | Number mode  
--- | --- | --- | ---  
In-cloud droplet mode | CD | $M_{CD}^{SU}, M_{CD}^{BC}, M_{CD}^{OC}, M_{CD}^{SS}, M_{CD}^{DU}$ | $N_{CD} = N_c$  
In-rain drop mode | RD | $M_{RD}^{SU}, M_{RD}^{BC}, M_{RD}^{OC}, M_{RD}^{SS}, M_{RD}^{DU}$ | $N_{RD} = N_r$  
In-ice crystal mode | IC | $M_{IC}^{SU}, M_{IC}^{BC}, M_{IC}^{OC}, M_{IC}^{SS}, M_{IC}^{DU}$ | $N_{IC} = N_i$  
In-snow flake mode | SF | $M_{SF}^{SU}, M_{SF}^{BC}, M_{SF}^{OC}, M_{SF}^{SS}, M_{SF}^{DU}$ | $N_{SF} = N_s$  
In-graupel mode | GR | $M_{GR}^{SU}, M_{GR}^{BC}, M_{GR}^{OC}, M_{GR}^{SS}, M_{GR}^{DU}$ | $N_{GR} = N_g$  

Table 2.2: Table of the five in-hydrometeor aerosol modes. $M_{\text{subst}}^{j}$ corresponds to the in-hydrometeor aerosol mass density for the mode $j \in \{CD, RD, IC, SF, GR\}$ and compound subst $\in \{SU, BC, OC, SS, DU\}$. (The number mode $N_j$ with $j \in \{CD, RD, IC, SF, GR\}$ is not a prognostic variable, but is diagnosed from the corresponding hydrometeor number density.)

dergo numerous cloud microphysical processes along with the hydrometeor. Upon evaporation and sublimation, the in-hydrometeor aerosol mass is released back to the atmosphere forming a newly generated aerosol particle. Due to falling hydrometeors reaching the ground, a part of the hydrometeor-borne aerosol mass is directly removed from the system, without being transferred back to the unactivated aerosol modes.

### 2.4 Aerosol-cloud interactions

In order to simulate aerosol-cloud interactions, the aerosol module has been coupled to the cloud microphysics scheme following the approach by Stier et al. (2005) as described in Muhlbauer and Lohmann (2008).

The new model version not only couples the aerosol module M7 and the cloud microphysics scheme, but additionally couples the five in-hydrometeor aerosol modes to both of them. This allows to simulate cycling of aerosol particles within clouds.

#### 2.4.1 Sulfur chemistry

The model considers two different origins of sulfate aerosol particles: direct emission from the earth surface and chemical formation within the atmosphere.

The chemical formation of sulfuric aerosol particles is represented by a gas-to-particle conversion from precursor gases or due to aqueous chemical reactions within cloud droplets. As precursor gases, we consider gas-phase sulfuric acid (H$_2$SO$_4$), sulfur dioxide (SO$_2$) and dimethyl sulfide (DMS) following the parameterization of the sulfur cycle by Feichter et al. (1996). The precursor gases are prognostic variables, whereas the oxidants OH, H$_2$O$_2$, NO$_2$ and O$_3$ are prescribed using climatological monthly means from the chemical transport model MOZART (Horowitz et al., 2003).
2.4. Aerosol-cloud interactions

Gas-phase chemistry

The model distinguishes between daytime and nighttime gaseous chemistry. During the day, gas-phase \( \text{H}_2\text{SO}_4 \) forms via an oxidation process of DMS and \( \text{SO}_2 \) with the OH-radical. At night, the oxidation of DMS by the nitrate radical (\( \text{NO}_3 \)) becomes most important. Regarding \( \text{NO}_3 \), we assume a steady state between the production and loss terms. Reactions and corresponding reaction rates are detailed in Feichter et al. (1996).

Gas-phase \( \text{H}_2\text{SO}_4 \) can then be transformed into sulfate aerosol particles via gas-to-particle conversion following the parameterization for sulfate nucleation by Vehkamäki et al. (2002). The parameterized nucleation rates of sulfuric acid and water are based on the classical nucleation theory.

Aqueous-phase chemistry in clouds

In cloud droplets, heterogeneous chemical reactions can lead to the formation of sulfate. We consider the formation of sulfate via the oxidation of \( \text{SO}_2 \) by \( \text{H}_2\text{O}_2 \) and \( \text{O}_3 \). Details on the reaction and the reactions coefficients can be found in Feichter et al. (1996).

Sulfate mass produced within cloud droplets was originally attributed to the accumulation and coarse mixed mode according to the mass ratio between the two modes as a crude representation of cloud processing. In the new model version including the in-hydrometeor aerosol modes, the produced sulfate aerosol mass is directly assigned to the in-cloud droplet mass mode \( M_{\text{CD}}^{SU} \).

2.4.2 Activation of cloud droplets

The activation of cloud droplets rests upon the activation of aerosol particles following the parameterization by Lin and Leaitch (1997). On the basis of the approaches by Lohmann (2002b), Muhlbauer and Lohmann (2008) and Zubler et al. (2011a) the number of newly activated cloud droplets depends on the number concentration of internally mixed, soluble aerosol particles larger than 35 nm (\( N_{c>35\,\text{nm}}^t \)) and the updraft vertical velocity \( w \) and is given by

\[
\frac{\partial N_c}{\partial t} = \max \left\{ \frac{1}{\Delta t} \left[ 0.1 \left( N_{c}^t \right)^{1.27} - N_{c}^{t-1} \right] , 0 \right\}
\]  

(2.6)

where

\[
N_c^t = \frac{w \, N_{c>35\,\text{nm}}^t}{w + \alpha \, N_{c>35\,\text{nm}}^{t-1}}
\]  

(2.7)

with \( \alpha = 0.023 \, \text{cm}^4\,\text{s}^{-1} \) and \( N_c \) the number of activated cloud droplets. As averaged over the entire gridbox, the simulated vertical velocity at a resolution of 12 \( \text{km} \) or more does not reach...
high enough values to realistically represent local updrafts. Therefore, the updraft velocity \( w \) is calculated as the sum of the grid-scale velocity \( w_g \) and a subgrid-scale turbulent contribution. Following the approach of Lohmann (2002b), the turbulent part depends on the turbulent kinetic energy (TKE) yielding for the updraft velocity

\[
w = w_g + c_{turb} \sqrt{TKE}, \quad c_{turb} = 1.33
\]  

(2.8)

\( N_{>35\text{ nm}} \) is the number concentration of possible CCN particles containing all internally mixed, soluble aerosols in the coarse \( (N_4) \) and accumulation mode \( (N_3) \) plus the number of mixed Aitken particles \( (N_2) \) with a wet radius larger than \( r_{crit} = 35 \text{ nm} \) such that

\[
N_{>35\text{ nm}} = N_4 + N_3 + \int_{\ln(r_{crit})}^{\infty} N_2(\ln r) d\ln r. \tag{2.9}
\]

Insoluble particles and particles smaller than 35 nm cannot act as CCN. When aerosol activation scavenging is considered, the number and mass of newly activated aerosol particles are removed from the M7 aerosol module. We assume that activation scavenging is progressing from the biggest to the smallest particle in each mode. Following Zubler et al. (2011a) the scavenged mass of each species in each mode corresponds to the lognormal tail of the size distribution and is calculated using a critical radius. Considering only scavenging processes without applying the aerosol processing scheme, activation scavenging implies the removal of the scavenged aerosols from the system. When the aerosol processing scheme is applied, it transfers the scavenged aerosol mass from the unactivated aerosol modes in the M7 module to the in-cloud droplet aerosol mode.

### 2.4.3 Heterogeneous freezing processes

Regarding heterogeneous nucleation of ice crystals, we account for freezing in the condensation / immersion and in the contact mode (Muhlbauer and Lohmann, 2009). Condensation freezing is implicitly included in the immersion freezing parameterization (Diehl et al., 2006; Lohmann and Diehl, 2006). In the following, the term immersion freezing will be used as a surrogate for both, immersion and condensation freezing modes. Experimental studies (Schaller and Fukuta, 1979; Welti et al., 2009) suggest coldest onset temperatures (between -30 and -20 °C) for deposition nucleation. It is believed the deposition mode becomes less important when water saturation is reached allowing for simultaneous freezing in the condensation mode. Due to the prevailing water saturation in mixed-phase clouds, we assume that deposition nucleation is less important in this cloud regime. In the parameterizations of immersion and contact
freezing, we consider black carbon (BC) and mineral dust (DU) as possible ice nuclei.

**Immersion freezing**

The parameterization of heterogeneous nucleation in the immersion mode is based on the stochastic hypothesis. Following this approach, heterogeneous freezing occurs, in analogy to homogeneous freezing, in supercooled cloud droplets due to random fluctuations amongst water molecules (Pruppacher and Klett, 1997). Thus, the freezing probability depends on the temperature and the droplet volume. Hence, larger droplets freeze at warmer temperatures than smaller droplets. Aerosol particles immersed in the droplet enhance the efficiency of random nucleation, but do not disturb its stochastic nature. Depending on their efficiency to serve as IN, they raise the probability of freezing for a fixed temperature.

The parameterization for heterogeneous freezing in the immersion mode is derived from Khain et al. (2000) and Seifert and Beheng (2006). The rate equation for the ice-crystal number density \( N_i \) is a function of the heterogeneous freezing rate \( J_{IFR}(T) \) such that

\[
\frac{\partial N_i}{\partial t} = -\frac{L_c}{\rho_w} J_{IFR}(T)
\]

(2.10)

Here, \( \rho_w \) is the density of water and \( L_c \) the mass density of cloud droplets. We assume conservation of number density \( \partial N_i / \partial t = -\partial N_c / \partial t \). Based on the parameterization of Diehl and Wurzler (2004) and Lohmann and Diehl (2006) the freezing rate can be determined by

\[
J_{IFR}(T) = a b \left\{ \exp \left[a \left(T_0 - T\right)\right] - 1 \right\} \frac{dT}{dt}, \quad T \leq T_0, \quad \frac{dT}{dt} < 0
\]

(2.11)

depending on the parameters \( a \) and \( b \), the temperature \( T \), the cooling rate \( dT/dt \) and the constant \( T_0 = 273.15 \) K. As described by Muhlbauer and Lohmann (2009) we explicitly account for different ice nucleation efficiencies of the different aerosol compounds by calculating the surface weighted averaged ice nucleation efficiency \( b \) as follows

\[
b = \sum_{\text{subst}} B_{\text{subst}} f^s_{\text{subst}, \text{subst}} \quad \text{subst} \in \{BC, DU\}
\]

(2.12)

Here, \( B_{\text{subst}} \) represents the material-specific, but size-dependent freezing efficiency of the aerosol component \( \text{subst} \) and \( f^s_{\text{subst}, \text{subst}} \) corresponds to the normalized surface fraction of the aerosol component \( \text{subst} \) which acts as an ice nucleus. For immersion freezing, we consider soluble mixed black carbon (BC) and mineral dust (DU) as efficient IN. The surface area fraction is considered since immersion freezing is assumed to be a surface area dependent process.
Table 2.3: Material specific coefficients for the parameterization of immersion freezing taken from table 2 in Diehl and Wurzler (2004), for soot (BC) and Montmorillonite (DU).

Contact freezing

Heterogeneous freezing in the contact mode is parameterized following the approaches of Young (1974a), Cotton et al. (1986) and Lohmann and Diehl (2006). The freezing process is assumed to be induced by collisions between supercooled water droplets and aerosol particles by Brownian diffusion (Muhlbauer and Lohmann, 2009).

\[
\frac{\partial N_i}{\partial t} = 4\pi \bar{r}_c N_c \sum_k D_k N_{k,\text{con}}, \quad k \in \{5, 6, 7\}
\]  

We therefore consider the aerosol Brownian diffusivity in air [m² s⁻¹]

\[
D_k = \frac{k_B TC}{6\pi \eta \bar{r}_k}
\]  

depending on the Boltzmann constant \( k_B = 1.38 \cdot 10^{-23} \text{ m}^2 \text{kg s}^2 \text{K}^{-1} \), the temperature \( T \), the Cunningham slip correction factor \( C \), the temperature dependent dynamic viscosity of air \( \eta \) and the dry count median radius \( \bar{r}_k \) for the aerosol mode \( k \). The Cunningham slip correction factor can be determined by

\[
C = 1 + 1.26 \left( \frac{\lambda}{\bar{r}_k} \right) \left( \frac{p_0}{p} \right) \left( \frac{T}{T_0} \right)
\]  

with the molecular mean free path length \( \lambda = 0.066 \mu\text{m} \) at standard condition \( p_0 = 1013.25 \text{ hPa} \) and \( T_0 = 273.15 \text{ K} \).

The Brownian diffusivity decreases with increasing particle size, leading to variations of the collision efficiency of several orders of magnitude between small and larger atmospheric aerosol particles. Thus, small particles like black carbon could be efficient IN, only due to their high collision efficiency. Assuming that contact IN need to be hydrophobic, and not already mixed or coated with any water soluble material, we limit the potential contact nuclei to the number concentration of insoluble aerosols. Following the experimental results by Gorbunov et al. (2001) we consider black carbon (BC) and mineral dust (DU) in the insoluble modes as potential nuclei for contact freezing. The sum of potential contact ice nuclei \( N_{k,\text{con}} \) in the aerosol mode \( k \) is
given by

\[ N_{k,\text{con}} = N_k \sum_{\text{subst}} \epsilon_{\text{subst}} f_{k,\text{subst}}, \quad \text{subst} \in \{BC, DU\} \]  

(2.16)

The freezing efficiency \( \epsilon_{\text{subst}} \) is calculated as a linear function of the temperature such that

\[ \epsilon_{\text{subst}} = c_1 T + c_0, \quad 0 \leq \epsilon_{\text{subst}} \leq 1 \]  

(2.17)

with the coefficients of the slope \( c_1 [K^{-1}] \) and the intercept \( c_0 \) depending on the aerosol type. The values are derived from laboratory measurements from Diehl et al. (2006) and given in table 2.4.3.

<table>
<thead>
<tr>
<th>Aerosol type</th>
<th>( c_1 [K^{-1}] )</th>
<th>( c_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Black Carbon (BC)</td>
<td>6.14 ( \cdot 10^{-2} )</td>
<td>57.30 ( \cdot 10^{-2} )</td>
</tr>
<tr>
<td>Mineral dust (DU)</td>
<td>10.14 ( \cdot 10^{-2} )</td>
<td>32.77 ( \cdot 10^{-2} )</td>
</tr>
</tbody>
</table>

Table 2.4: Material specific coefficients for the parameterization of contact freezing taken from table 3 in Diehl et al. (2006), for soot (BC) and Montmorillonite (DU).

2.4.4 In-cloud collision scavenging

The model further considers collisions between aerosol particles and hydrometeors. In-cloud collision scavenging refers to collision-coalescence of aerosol particles with cloud droplets or ice crystals. The collision rate \( Q_{\text{hydro},k} \) between hydrometeors class \( \text{hydro} \) and aerosol particles of the mode \( k \) can be calculated as follows

\[ Q_{\text{hydro},k} = K_{\text{hydro},k} N_{\text{hydro}} N_k \]  

(2.18)

It depends on the prescribed collision kernel \( K_{\text{hydro},k} \), the hydrometeor number concentration \( N_{\text{hydro}} \) with \( \text{hydro} \in \{c, i\} \) and the aerosol number concentration of the mode \( k \in [1, 7] \).

For cloud droplets, the collision kernels \( K_{c,k} \) are estimated from Young (1974b) for growing droplets following the approach of Hoose et al. (2008a). We assume the count median radii for the aerosol modes and an average cloud droplet radius of 10 \( \mu m \). For ice crystals, the shape dependency is neglected and the values for the collision kernels \( K_{i,k} \) are estimated from Young (1974b) for an average crystal radius of 15 \( \mu m \). Variations of the actual diameters are neglected. The obtained collision kernels are listed in Table 2.5.
Chapter 2. Model description

### 2.5 Aerosol mode

<table>
<thead>
<tr>
<th>Aerosol mode</th>
<th>( K_{c,k} )</th>
<th>( K_{i,k} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nucleation mixed mode</td>
<td>( 2.5 \cdot 10^{-12} )</td>
<td>( 5.0 \cdot 10^{-11} )</td>
</tr>
<tr>
<td>Aitken mixed mode</td>
<td>( 2.5 \cdot 10^{-12} )</td>
<td>( 5.0 \cdot 10^{-11} )</td>
</tr>
<tr>
<td>Accumulation mixed mode</td>
<td>( 2.5 \cdot 10^{-14} )</td>
<td>( 2.0 \cdot 10^{-12} )</td>
</tr>
<tr>
<td>Coarse mixed mode</td>
<td>0</td>
<td>( 2.0 \cdot 10^{-13} )</td>
</tr>
<tr>
<td>Aitken insoluble mode</td>
<td>( 2.5 \cdot 10^{-12} )</td>
<td>( 5.0 \cdot 10^{-11} )</td>
</tr>
<tr>
<td>Accumulation insoluble mode</td>
<td>( 2.5 \cdot 10^{-12} )</td>
<td>( 2.0 \cdot 10^{-12} )</td>
</tr>
<tr>
<td>Coarse insoluble mode</td>
<td>( 2.5 \cdot 10^{-14} )</td>
<td>( 2.0 \cdot 10^{-13} )</td>
</tr>
</tbody>
</table>

**Table 2.5:** Collision kernels \( K_{c,k} \) for aerosol-droplet collisions and \( K_{i,k} \) for aerosol-crystal collisions for the aerosol mode \( k \in \{1, 7\} \) given in \( \text{m}^3 \text{s}^{-1} \) after Young (1974b) obtained for a droplet radius of 10 \( \mu \text{m} \) and a crystal radius of 15 \( \mu \text{m} \) and the average aerosol size of the corresponding mode.

### 2.4.5 Below-cloud scavenging

Below cloud scavenging denotes the scavenging of aerosol particles by sedimenting hydrometeors. We consider below-cloud scavenging by rain and snow, omitting graupel because of its rare occurrence and its large sedimentation velocity leading to a short residence time in the atmosphere.

Based on the equations given by Zhao and Zheng (2006) and Croft et al. (2009), the scavenging coefficient \( \Lambda(D_a) \) is calculated for rain and snow as described by Zubler et al. (2011a) such that

\[
\Lambda(D_a) = \int_{D_{h,min}}^{D_{h,max}} \frac{\pi D_h^2}{4} U_t(D_h) \cdot E(D_a, D_h) \cdot N_h(D_h) \ dD_h
\]  

(2.19)

where \( D_a \) represents the aerosol particles diameter, \( D_h \) is the hydrometeor (rain or snow) diameter, \( D_{h,max} \) and \( D_{h,min} \) denote the maximum and minimum hydrometeor size within the cloud microphysics scheme, \( U_t \) corresponds to the terminal fall velocity, \( N_h(D_h) \) represents the number density size distribution of the hydrometeor class. The collection efficiency \( E(D_a, D_h) \) is composed of the scavenging efficiencies due to Brownian motion, interception and inertial impaction. Although thermo- and diffusiophoretic forces play an important role for the removal of accumulation mode aerosol particles, they are not accounted for in this approach for the sake of simplicity. The mass scavenging coefficient \( \Lambda_m \) and the number scavenging coefficient \( \Lambda_n \) can be calculated by integration of \( \Lambda(D_a) \) over the aerosol size distribution \( N(D_a) \) such that

\[
\Lambda_m = \int_0^\infty \Lambda(D_a) \cdot D_a^3 \cdot N(D_a) \ dD_a \int_0^\infty D_a^3 \cdot N(D_a) \ dD_a
\]

(2.20)
and

$$\Lambda_n = \frac{\int_0^\infty \Lambda(D_a) \, N(D_a) \, dD_a}{\int_0^\infty N(D_a) \, dD_a}$$

(2.21)

In Zubler et al. (2011a), the rate change of a tracer mixing ratio $\frac{dM}{dt}$ due scavenging by rain (or snow) depends also on the fraction of precipitating rain (or snow) $f_{\text{precip}}$ such that

$$\frac{dM}{dt} = -M \, f_{\text{precip}} \, \Lambda_m.$$  

(2.22)

A corresponding equation also exists for the aerosol number tracers.

In the new aerosol processing scheme, the scavenged aerosol mass and number are independent of the fraction of precipitating rain (or snow), as every collision-coalescence between rain (or snow) and aerosol particles leads to a transfer of aerosol mass from the unactivated aerosol mode of the M7 module to the corresponding in-rain (or in-snow) aerosol mode $M_{\text{RD}}$ (or $M_{\text{SF}}$) with $\text{subst} \in \{\text{SU, BC, OC, SS, DU}\}$.

### 2.4.6 Evaporation / sublimation of hydrometeors

Aerosol particles are regenerated following complete evaporation or sublimation of cloud particles. A detailed description of the parameterization of the evaporation and sublimation processes within the microphysical scheme is given in Seifert and Beheng (2006). Here, we focus on the implications of hydrometeor evaporation and / or sublimation for the aerosol population.

Following the homogeneous mixing assumption, we consider residual aerosol particles of evaporation and sublimation to contain an internal mixture of all foreign material in the hydrometeor. Due to sulfate production in the aqueous phase, all processed particles contain some sulfate mass and therefore can be attributed to the unactivated mixed / soluble modes. Based on wind tunnel studies of evaporating drops by Mitra et al. (1992) one can assume that the released particles do not break up during drop evaporation. We assume the same for the residuals of sublimating ice hydrometeors.

The model necessarily has to make assumptions regarding the size distributions of the regenerated aerosol particles. Representing the newly formed aerosol particles by a lognormal function, a gamma function or a single size bin with the diameter equal to the volume-mean diameter of all the regenerated particles Yin et al. (2005) found that the impact of different size
distributions on the number concentration of CCN lies within ± 1% for their case study. Adopting lognormal size distributions for the regenerated aerosol particles, we calculate the dry count median radius following Hoose et al. (2008a) such that

\[
\bar{r}_j = \sqrt[3]{\frac{3}{4\pi} \frac{V_j^{\text{tot}}}{N_j} \exp \left( -\frac{3 \ln^2 \sigma_j}{2} \right)}, \quad j \in \{CD, IC, RD, SF, GR\}
\] (2.23)

with \(N_j\) number density of potentially released aerosol particles (equal to the number density of corresponding hydrometeors), \(\sigma_j\) the geometric standard deviation of the size distribution being fixed to a value of 1.59 for released aerosol particles smaller than 0.5 \(\mu\)m and to the values of the coarse mode (= 2.0) for larger particles. \(V_j^{\text{tot}}\) representing the total in-hydrometeor volume of aerosol mass within the grid box is given by

\[
V_j^{\text{tot}} = \sum_l \frac{M_l^{\text{subst}}}{\rho_{\text{subst}}}, \quad \text{subst} \in \{SU, BC, OC, SS, DU\}
\] (2.24)

The dry densities of the compounds are given by \(\rho_{SU} = 1841 \text{ kg m}^{-3}\), \(\rho_{BC} = \rho_{OC} = 2000 \text{ kg m}^{-3}\), \(\rho_{SS} = 2165 \text{ kg m}^{-3}\) and \(\rho_{DU} = 2650 \text{ kg m}^{-3}\). The released aerosol particles are attributed to the unactivated internally mixed / soluble mode with \(\bar{r}_m\) falling in its size range (table 2.1).
Chapter 3

Microphysical processing of aerosol particles in orographic clouds

S. Pousse-Nottelmann $^{1,3}$, E. M. Zubler $^2$ and U. Lohmann $^{1,3}$


An explicit and detailed treatment of cloud-borne particles allowing for the consideration of aerosol cycling in clouds has been implemented into COSMO-Model, the regional weather forecast and climate model of the Consortium for Small-scale Modeling (COSMO). The effects of aerosol scavenging, cloud microphysical processing and regeneration upon cloud evaporation on the aerosol population and on subsequent cloud formation are investigated. For this, two-dimensional idealized simulations of moist flow over two bell-shaped mountains were carried out varying the treatment of aerosol scavenging and regeneration processes for a warm-phase and a mixed-phase orographic cloud.

The results allowed us to identify different aerosol cycling mechanisms. In the simulated non-precipitating warm-phase cloud, aerosol mass is incorporated into cloud droplets by activation scavenging and released back to the atmosphere upon cloud droplet evaporation. In the mixed-phase cloud, a first cycle comprises cloud droplet activation and evaporation via the Wegener-Bergeron-Findeisen (WBF) process. A second cycle includes below-cloud scavenging by precipitating snow particles and snow sublimation and is connected to the first cycle via the riming process which transfers aerosol mass from cloud droplets to snowflakes. In the simulated mixed-phase cloud, only a negligible part of the total aerosol mass is incorporated into ice crystals. Sedimenting snowflakes reaching the surface remove aerosol mass from the atmosphere. The results show that aerosol processing and regeneration lead to a vertical redistribution of aerosol mass and number. Thereby, the processes impact the total aerosol number and mass and additionally alter the shape of the aerosol size distributions by enhancing the internally mixed/soluble Aitken and accumulation mode and generating coarse-mode particles. Concerning subsequent cloud formation at the second mountain, accounting for aerosol processing and regeneration increases the cloud droplet number concentration with possible implications for the ice crystal number concentration.

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Parts of this publication are based on chapter 2 and thus repeated here.
Chapter 3. Microphysical processing of aerosol particles in orographic clouds

3.1 Introduction

Orography has an important influence on precipitation formation and can be a key factor in hydrology, ecology and climate on the local scale (Smith et al., 2005; Saleeby et al., 2009). When air rises over sloped terrain, it cools. Above the lifting condensation level, the air becomes supersaturated with respect to water allowing for the formation of cloud droplets by condensation of water vapor onto cloud condensation nuclei (CCN). In such a warm-phase cloud, precipitation can form via condensational growth and collision–coalescence of cloud droplets. When temperatures fall below the freezing level, ice crystal formation is triggered by ice nuclei (IN), transforming the liquid cloud into a mixed-phase cloud. Due to the difference in saturation vapor pressure over ice and over water, ice crystals can grow rapidly at the expense of evaporating cloud droplets when the vapor pressure is below water saturation. This process, known as the Wegener–Bergeron–Findeisen (WBF) process (Wegener, 1911; Bergeron, 1935; Findeisen, 1938), can lead to a rapid glaciation of a mixed-phase cloud at temperatures between $-38$ and $0^\circ C$. Below $-38^\circ C$ the droplets start to freeze homogeneously.

Ice microphysical processes are of major interest, as 70% of tropical precipitation forms via the ice phase (Lau and Wu, 2003). On the regional scale, heavy orographic precipitation events in the Alps have also been associated with ice-phase processes (Pujol et al., 2005), indicating the importance of the cold phase for precipitation formation.

Aerosol particles impact the cloud microphysical processes directly by serving as cloud condensation nuclei for the formation of cloud droplets and by initiating ice nucleation. They influence cloud development and properties and ultimately precipitation formation. However, the quantification of the aerosol effect on clouds remains challenging and represents one of the largest uncertainties in the climate system (Boucher et al., 2013).

Enhanced aerosol concentrations result in an increased number concentration of cloud droplets, shifting the droplet size spectrum towards smaller radii for a constant liquid water content (Twomey et al., 1974). Due to the reduced collision efficiencies of smaller droplets, the formation of rain is delayed and the cloud may persist for a longer time period (Albrecht, 1989).

In idealized simulations of purely warm-phase orographic precipitation, Muhlbauer and Lohmann (2008) observed that increased atmospheric aerosol concentrations lead to more but smaller cloud droplets producing a shift in precipitation towards the leeward side of the mountain (spillover effect). An investigation of orographic precipitation over the Sierra Nevada showed that polluted continental aerosol concentrations in comparison to clean marine aerosol
conditions lead to a precipitation reduction on the upslope side of the mountains, decreasing the overall precipitation by about 30% (Lynn et al., 2007).

If ice processes are involved, the difference between clouds formed on marine vs. continental aerosol concentrations is even more pronounced (Lynn et al., 2007). Indeed, within mixed-phase cloud regimes, a decrease of the mean cloud droplet radius also implies a lessening of the riming efficiency between droplets and ice particles due to reduced collision efficiencies of smaller droplets (Lohmann, 2004). Saleeby et al. (2009) showed that in mixed-phase orographic clouds along the Park Range barrier in Colorado, enhanced CCN number concentrations modify the droplet distribution significantly, leading to a substantial inhibition of the riming process. At the same time, the WBF process is enhanced under polluted conditions because of the greater surface area exposure of more but smaller cloud droplets, largely compensating for the loss in snow growth by the reduced riming process (Saleeby et al., 2013).

Another important characteristic is the ice-nucleation ability of certain aerosol types. Depending on the composition and mixing state, a part of the aerosol population may potentially serve as ice nuclei and trigger heterogeneous freezing. Depending on the temperature and supersaturation with respect to water, different freezing modes have been identified (Vali, 1985). Zubler et al. (2011b) showed that the effect of an increased aerosol number concentration on orographic mixed-phase clouds strongly depends on the presence of the ice phase and particularly the heterogeneous freezing of droplets. In cold simulations, with aerosol particles acting as CCN and as IN in the condensation/immersion freezing mode, the reduction of precipitation formation via the liquid phase due to smaller droplets is partly compensated by a glaciation of the clouds. The processes which are most affected by increased aerosol loadings are coalescence and riming (Muhlbauer and Lohmann, 2009), which are important pathways for precipitation formation.

Muhlbauer and Lohmann (2009) investigated the impact of dust and anthropogenic black carbon anomalies as potential ice nuclei on orographic mixed-phase clouds and pointed out the crucial role of the assumed aerosol properties. Due to the different onset temperatures and efficiencies of the different freezing modes, the ice phase may vary according to the dominant freezing process impacting riming rates and thus precipitation formation.

During their residence time in the atmosphere, aerosol particles undergo various modifications which may affect their ability to serve as CCN and IN. Thereby, aerosol processing within clouds plays a crucial role, as the aerosol mass, composition and mixing state can be altered. Due to activation, nucleation and collision–coalescence processes aerosol particles are incorporated
into hydrometeors. Processes like autoconversion, accretion, aggregation, freezing, melting, riming and self-collection transfer aerosol mass and number between the different hydrometeor classes. Aqueous-phase chemistry within droplets can lead to the formation of new aerosol mass. Finally, wet deposition, sedimentation and scavenging processes lead to a removal of aerosol mass from the atmosphere. However, a substantial fraction of hydrometeors evaporate/sublimate, releasing newly formed aerosol particles to the atmosphere with different size, composition and mixing state compared to the original ones. Recent investigations concerning the impact of aerosol solubility and recycled aerosol particles on orographic cloud formation have been conducted by Xue et al. (2010, 2012). They explicitly account for the release of aerosol particles upon droplet and rain evaporation, replenishing between one-third and two-thirds of the scavenged aerosol particles. In their model configuration, the CCN number concentration explicitly depends on the number concentration and properties of the background and regenerated aerosol particles. In idealized 2-D simulations of warm-phase clouds over two bell-shaped mountains, recycled aerosol particles enhance the cloud droplet number concentration and thus reduce precipitation formation at the second mountain (Xue et al., 2010). In mixed-phase clouds, regenerated aerosol particles were found to inhibit the riming process by changing the droplet size distribution (Xue et al., 2012).

In the atmosphere, however, cloud droplet activation and heterogeneous freezing processes may be affected by the release of processed aerosol particles. The ice-nucleation efficiency of aerosol particles in the different freezing modes depends on their size, composition and mixing state, which may be altered during cloud processing. The present study therefore explicitly considers both the droplet and ice nucleating ability of certain aerosol particles and their release back into the atmosphere upon evaporation/sublimation.

To better understand the role of aerosol–cloud interactions and especially the role of regenerated aerosol particles, COSMO-Model, the regional weather forecast and climate model currently developed and maintained by the Consortium for Small-scale Modeling (COSMO), has been extended in a detailed treatment of hydrometeor-borne aerosol particles following the approach by Hoose et al. (2008a). The new scheme allows for the simulation of cloud cycling of aerosols by tracing them even when scavenged into hydrometeors.

The paper is structured as follows: in Sect. 3.2 we describe the employed cloud and aerosol microphysical parameterizations with a focus on aerosol–cloud interactions. In Sect. 3.3 we introduce the simulation setup before presenting the results for warm-phase clouds in Sect. 3.4.1 and mixed-phase clouds in Sect. 3.4.2. In Sect. 3.5 we end with a summary of our results and conclusions.
3.2 Numerical model description

This study makes use of COSMO-Model (http://www.cosmo-model.org), a non-hydrostatic, fully compressible weather prediction and climate model. This limited-area mesoscale atmospheric prediction model integrates the elastic hydro-thermodynamical equations on a rotated Arakawa C-grid with a time-splitting approach following a split-explicit third-order Runge–Kutta scheme in combination with a fifth-order upstream horizontal advection scheme (Doms and Schättler, 2002; Steppeler et al., 2003). The aerosol and moisture variables are advected by the second-order positive-definite Bott scheme (Bott, 1989).

3.2.1 Cloud microphysics

Cloud microphysical processes are calculated within the two-moment bulk cloud microphysics scheme for water and ice clouds of Seifert and Beheng (2006). The scheme comprises prognostic equations for the mass and number densities of five different hydrometeor classes: cloud droplets, ice crystals, raindrops, snowflakes and graupel. Warm-phase cloud processes include activation of cloud droplets, condensational growth and evaporation of cloud droplets, autoconversion (formation of rain by coagulating cloud droplets) and accretion (growth of raindrops by collection of cloud droplets), self-collection (mutual coagulation of cloud droplets/raindrops, remaining in the same drop category), evaporation of rain and collisional break-up of large raindrops. In colder regimes, the scheme also accounts for homogeneous and heterogeneous freezing, diffusional growth of ice crystals, aggregation (formation of snow by coagulating ice crystals), self-collection (mutual coagulation of ice/snow/graupel, remaining in the same hydrometeor category), riming (coagulation between the liquid and the ice phase), melting and sublimation and related secondary processes. The Wegener–Bergeron–Findeisen process is represented implicitly in the model because in an air parcel, which is supersaturated with respect to ice but subsaturated with respect to water, all condensate evaporates and the ice crystals grow by water diffusion. The parameterization of hydrometeor sedimentation is based on mass- and number-weighted mean fall velocities.

3.2.2 Aerosol microphysics

The M7 aerosol module

The cloud microphysics scheme is coupled to the M7 aerosol module (Vignati et al., 2004; Muhlbauer and Lohmann, 2008, 2009; Zubler et al., 2011a) which is also used within the framework of the general circulation model system ECMWF Hamburg Model (ECHAM5-HAM) (Stier
et al., 2005). The M7 aerosol module describes the aerosol population as a superposition of seven lognormal size distributions of the form

\[ N(\ln r) = \sum_{k=1}^{7} \frac{N_k}{\sqrt{2\pi} \ln \sigma_k} \exp \left[ -\left( \frac{\ln r - \ln r_k}{\sqrt{2} \ln \sigma_k} \right)^2 \right]. \] (3.1)

Here, \( N_k \) represents the aerosol number concentration of the \( k \)th aerosol mode with \( k \in [1, 7] \) and \( \sigma_k \) is the corresponding geometric standard deviation. In M7, the aerosol number and mass concentrations are prognostic variables whereas the standard deviations are kept constant.

The module distinguishes between four internally mixed, soluble modes (nucleation, Aitken, accumulation, and coarse mode), containing both soluble and insoluble compounds, and three insoluble modes (Aitken, accumulation, and coarse mode), which are characterized by low water solubility. It accounts for five different aerosol compounds: sulfate (SU), carbonaceous aerosols (black carbon BC, organic carbon OC), sea salt (SS) and mineral dust (DU) (Table 3.1).

<table>
<thead>
<tr>
<th>Aerosol mode</th>
<th>Size range [( \mu \text{m} )]</th>
<th>Soluble/externally mixed mode</th>
<th>Insoluble modes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nucleation</td>
<td>&lt; 0.005</td>
<td>( N_1, M_1^{SU} )</td>
<td></td>
</tr>
<tr>
<td>Aitken</td>
<td>0.005 – 0.05</td>
<td>( N_2, M_2^{SU}, M_2^{BC}, M_2^{OC} )</td>
<td>( N_5, M_5^{BC}, M_5^{OC} )</td>
</tr>
<tr>
<td>Accumulation</td>
<td>0.05 – 0.5</td>
<td>( N_3, M_3^{SU}, M_3^{BC}, M_3^{OC}, M_3^{SS}, M_3^{DU} )</td>
<td>( N_6, M_6^{DU} )</td>
</tr>
<tr>
<td>Coarse</td>
<td>&gt; 0.5</td>
<td>( N_4, M_4^{SU}, M_4^{BC}, M_4^{OC}, M_4^{SS}, M_4^{DU} )</td>
<td>( N_7, M_7^{DU} )</td>
</tr>
</tbody>
</table>

**Table 3.1:** Table of the different aerosol modes of the aerosol module M7. The size range of each mode is given for the count median radius \( r_k \). \( N_k \) and \( M_k^X \) correspond to the aerosol number and mass density for the mode \( k \in [1,7] \) and compound \( X \in \{ SU, BC, OC, SS, DU \} \) respectively. Adapted from Vignati et al. (2004).

Due to sulfate coating or coagulation, insoluble particles, containing externally mixed freshly formed (black) carbon and dust, can be transferred to the internally mixed modes. The aerosol scheme accounts for nucleation of gas-phase sulfuric acid (Vehkamäki et al., 2002), condensation of sulfuric acid on pre-existing aerosol particles, coating of insoluble aerosol particles with sulfuric acid, inter- and intramodal coagulation and water vapor uptake. The M7 aerosol module does not account for nitrate and ammonium aerosol components as it does not include a treatment of the thermodynamic equilibration of these semi-volatile aerosol compounds with the gas phase. However, these constituents may represent an important part of the aerosol population and modify the surface properties of other aerosol particles due to coatings increasing aerosol growth and activation to cloud droplets. This work represents the aerosol population with the modal approach which is computationally cheaper than the sectional approach and allows one to conduct 3-D long-term simulations. Comparing the modal and sectional approach
in a global 3-D model, Mann et al. (2012) found that differences between the two approaches were less than model–observation differences. However, they underlined that the size-resolved aerosol properties in modal schemes need to be benchmarked and improved against sectional schemes and observations. Applying a two-moment aerosol bulk microphysics scheme with an explicit treatment of aerosol activation and scaled rates for the other microphysical processes, Lebo and Morrison (2013) obtained similar results between the modal and the sectional approach. The explicit parameterizations of aerosol activation and below-cloud scavenging processes in our model were evaluated against observations by Zubler et al. (2011a).

The aerosol processing scheme

To simulate aerosol processing in clouds, five new aerosol modes corresponding to the five prognostic hydrometeor classes (in-cloud droplet (CD), in-ice crystal (IC), in-raindrop (RD), in-snowflake (SF) and in-graupel (GR) aerosol mode; Table 3.2) are introduced into the model. Following the approach of Hoose et al. (2008a), these new modes contain hydrometeor-borne aerosol mass, potentially consisting of a contribution of all five aerosol components (SU, BC, OC, SS, DU). The number densities of hydrometeor-borne aerosol particles are not prognostic variables, but diagnosed from the corresponding hydrometeor number density during evaporation and sublimation.

<table>
<thead>
<tr>
<th>Aerosol mode</th>
<th>Abbrev.</th>
<th>Mass modes</th>
<th>Number mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>In-cloud droplet mode</td>
<td>CD</td>
<td>$M_{CD}^{SU}$, $M_{CD}^{BC}$, $M_{CD}^{OC}$, $M_{CD}^{SS}$, $M_{CD}^{DU}$</td>
<td>$N_{CD} = N_c$</td>
</tr>
<tr>
<td>In-raindrop mode</td>
<td>RD</td>
<td>$M_{RD}^{SU}$, $M_{RD}^{BC}$, $M_{RD}^{OC}$, $M_{RD}^{SS}$, $M_{RD}^{DU}$</td>
<td>$N_{RD} = N_r$</td>
</tr>
<tr>
<td>In-ice crystal mode</td>
<td>IC</td>
<td>$M_{IC}^{SU}$, $M_{IC}^{BC}$, $M_{IC}^{OC}$, $M_{IC}^{SS}$, $M_{IC}^{DU}$</td>
<td>$N_{IC} = N_i$</td>
</tr>
<tr>
<td>In-snowflake mode</td>
<td>SF</td>
<td>$M_{SF}^{SU}$, $M_{SF}^{BC}$, $M_{SF}^{OC}$, $M_{SF}^{SS}$, $M_{SF}^{DU}$</td>
<td>$N_{SF} = N_s$</td>
</tr>
<tr>
<td>In-graupel mode</td>
<td>GR</td>
<td>$M_{GR}^{SU}$, $M_{GR}^{BC}$, $M_{GR}^{OC}$, $M_{GR}^{SS}$, $M_{GR}^{DU}$</td>
<td>$N_{GR} = N_g$</td>
</tr>
</tbody>
</table>

Table 3.2: Table of the five in-hydrometeor aerosol modes. $M_{j}^{X}$ corresponds to the in-hydrometeor aerosol mass density for the mode $j \in \{CD, RD, IC, SF, GR\}$ and compound $X \in \{SU, BC, OC, SS, DU\}$. The number mode $N_{j}$ with $j \in \{CD, RD, IC, SF, GR\}$ is not a prognostic variable, but is diagnosed during evaporation and sublimation from the corresponding hydrometeor number density.

The new modes are not limited by any specific size range. Their median radii are calculated directly upon evaporation/sublimation (Eq. 3.5). Aerosol activation scavenging, collision/impaction scavenging as well as sulfate production within droplets lead to an increase of the in-hydrometeor aerosol mass. In the standard version of the model as described by Muhlbauer and Lohmann (2008), the M7 aerosol module comprises the total aerosol population, including interstitial particles within the cloud, unactivated particles in cloud free air and activated in-hydrometeor aerosols. In the new model version presented here, only interstitial or...
unactivated aerosol particles are assigned to the M7 module. All material which is taken up by a hydrometeor is transferred to the new modes CD, IC, RD, SF and GR.

Aerosol particles are incorporated into hydrometeors due to the different scavenging processes, including activation of aerosol particles, in-cloud impaction scavenging and below-cloud scavenging by precipitating hydrometeors. Once inside a hydrometeor, the aerosol particles undergo the cloud microphysical processes along with the hydrometeors. Upon evaporation and sublimation, the in-hydrometeor aerosol mass is released to the atmosphere forming a newly generated aerosol particle. To allow for coagulation, sedimentation, activation and scavenging of the processed aerosol particles together with the unactivated background aerosol, processed and background aerosol are summed up loosing valuable information about their specific size distributions and composition. When falling hydrometeors reach the ground, the hydrometeor-borne aerosol mass is directly removed from the system. These processes and the corresponding transfer of aerosol mass are illustrated in Fig. 3.1.

**Figure 3.1:** Microphysical transfer processes between the different aerosol modes (coll represents in-cloud and below-cloud collision scavenging, act represents activation scavenging, frz represents freezing, melt represents melting, auto represents autoconversion, acc represents accretion, agg represents aggregation, rim represents riming, enh. melt represents enhanced melting, ice multi represents ice multiplication, self collec represents self-collection, evap represents evaporation, sub represents sublimation).

### 3.2.3 Aerosol–cloud interactions

In order to simulate aerosol–cloud interactions, the aerosol module has been coupled to the cloud microphysics scheme following the approach by Stier et al. (2005) as described in
3.2. Numerical model description

Muhlbauer and Lohmann (2008). In this standard version of the model, the aerosol number concentrations influence the cloud properties, but are not affected themselves by the clouds.

The new aerosol processing scheme tracks aerosol particles even when incorporated into hydrometeors. The aerosol module M7, the cloud microphysics scheme, and the five in-hydrometeor aerosol modes are directly coupled in order to allow for the simulation of cloud cycling of aerosol particles. This enables to consider the interactions between aerosols and clouds in a complete way.

Activation of cloud droplets

In the present model version the activation of cloud droplets is parameterized according to the approach by Lin and Leaitch (1997) following the works by Muhlbauer and Lohmann (2008) and Zubler et al. (2011a). The number of newly activated cloud droplets is based on the number concentration of soluble/mixed aerosol particles larger than 35 nm \( (N_{>35\text{ nm}}) \) and the updraft vertical velocity \( w \) and is given by

\[
\frac{\partial N_c}{\partial t} = \max \left\{ \frac{1}{\Delta t} \left[ 0.1 \left( \frac{w N_{>35\text{ nm}}}{w + \alpha N_{>35\text{ nm}}} \right)^{1.27} - N_c^{t-1} \right], 0 \right\}, \tag{3.2}
\]

with \( \alpha = 0.023 \text{ cm}^4 \text{s}^{-1} \) and \( N_c \) the number of activated cloud droplets. The simulated vertical velocity may not reach high enough values to realistically represent local updrafts when averaged over the entire grid box, especially in simulations with lower horizontal resolution. Therefore, the updraft velocity \( w \) is calculated as the sum of the grid-scale velocity \( w_g \) and a subgrid-scale turbulent contribution. Following the approach of Lohmann (2002b), the turbulent part depends on the turbulent kinetic energy (TKE) yielding for the updraft velocity

\[
w = w_g + c_{\text{turb}} \sqrt{\text{TKE}}, \quad c_{\text{turb}} = 1.33. \tag{3.3}
\]

To be consistent with the standard parameterization of cloud droplet activation in the model we maintain this calculation of the updraft velocity in our simulations though not imperative at a horizontal resolution of 2 km.

\( N_{>35\text{ nm}} \) is the number concentration of possible CCN particles containing all soluble/mixed aerosols in the coarse \( (N_4) \) and accumulation modes \( (N_3) \) plus the number of mixed Aitken-mode particles \( (N_2) \) with a wet radius larger than \( r_{\text{crit}} = 35 \text{ nm} \) such that

\[
N_{>35\text{ nm}} = N_4 + N_3 + \int_{\ln(r_{\text{cri}})}^{\infty} N_2(\ln r) \, d \ln r. \tag{3.4}
\]
Insoluble particles and particles smaller than 35 nm cannot act as cloud condensation nuclei. When aerosol activation scavenging is considered, the number and mass of newly activated aerosol particles are removed from the M7 aerosol module. We assume that activation scavenging progresses from the biggest to the smallest particle in each mode. Following Zubler et al. (2011a) the scavenged mass of each species in each mode corresponds to the lognormal tail of the size distribution and is calculated using a critical radius. Scavenging thus causes a deviation from the log-normal aerosol modes that is lost when the new log-normal size distribution with reduced number concentration and mode radius is calculated. Considering only scavenging processes without applying the aerosol processing scheme, activation scavenging implies the removal of the scavenged aerosols from the system. When the aerosol processing scheme is applied, it transfers the scavenged aerosol mass from the unactivated aerosol modes in the M7 module to the in-cloud droplet aerosol mode. In the standard version of the model, no activation scavenging is considered.

**Heterogeneous freezing processes**

Regarding heterogeneous nucleation of ice crystals, the model accounts for freezing in condensation/immersion and in contact mode (Muhlbauer and Lohmann, 2009), neglecting deposition nucleation due to the prevailing water saturation in mixed-phase clouds. The parameterization of immersion-mode freezing derived from Diehl and Wurzler (2004) and Lohmann and Diehl (2006) is based on the stochastic hypothesis considering that immersed aerosol particles can enhance the efficiency of random ice nucleation in a droplet, but do not disturb its stochastic nature. For immersion freezing, we consider soluble mixed black carbon (BC) and mineral dust (DU) as efficient ice nuclei dependent on a material-specific, size-dependent freezing efficiency and on the surface area fraction (Muhlbauer and Lohmann, 2009). Heterogeneous freezing in the contact mode is parameterized following the approaches of Young (1974a), Cotton et al. (1986) and Lohmann and Diehl (2006). The freezing process is assumed to be induced by collisions between supercooled water droplets and aerosol particles by Brownian diffusion (Muhlbauer and Lohmann, 2009). We consider insoluble (hydrophobic) black carbon (BC) and mineral dust (DU) as potential nuclei for contact freezing (Diehl et al., 2006). The Brownian diffusivity decreases with increasing particle size, leading to variations of the collision efficiency of several orders of magnitude between small and larger atmospheric aerosol particles.

**Collision scavenging**

The new model version further considers scavenging of aerosol particles by collision with hydrometeors. In-cloud collision scavenging refers to collision–coalescence of aerosol particles
3.2. Numerical model description

with cloud droplets or ice crystals. The parameterization of the collision rate depends on pre-
scribed collision kernels estimated by Young (1974b), the aerosol number concentration and
the cloud droplet (ice crystal) number concentration. Collision scavenging by sedimenting rain
or snow is called below-cloud scavenging. The parametrization is based on the scavenging
coefficient following the equations in Zhao and Zheng (2006) and Croft et al. (2009), as de-
scribed by Zubler et al. (2011a). In order to consider only the final removal of aerosol particles
from the atmosphere by surface reaching hydrometeors, Zubler et al. (2011a) multiplied the
rate change of the tracer mass and number densities due to scavenging by rain (or snow) by
the fraction of precipitating rain (or snow) reaching the surface. Scavenging by falling graupel
is omitted. In the new aerosol processing scheme, all precipitating raindrops and snowflakes,
not only surface-reaching hydrometeors, may scavenge aerosol particles. In this scheme, the
scavenging coefficients are thus independent from the fraction of surface reaching precipitation.
The scavenged aerosol mass is attributed to the corresponding in-hydrometeor aerosol-mode
\[ M_X \] for in-cloud collision scavenging and \[ M_{RD}^X \] or \[ M_{SF}^X \] for below-cloud scavenging with
\( X \in \{SU, BC, OC, SS, DU\} \). The standard version of the model does not account for collision
scavenging.

Evaporation/sublimation of hydrometeors

During evaporation and sublimation of hydrometeors, aerosol particles are released back to
the atmosphere. In-hydrometeor aerosol mass and the corresponding hydrometeors are trans-
ported the same way. Within one model time step, hydrometeors may also fall through several
vertical model layers as the sedimentation flux calculations are done on a smaller time step.
We scale the in-hydrometeor sedimentation flux according to the sedimentation flux of the cor-
responding hydrometeor. Therefore, the locations of evaporation/sublimation and the release of
a new aerosol particles upon evaporation/sublimation coincide. The emitted aerosol particles
are assumed to not break up (Mitra et al., 1992) and we presume that they contain a mixture of
all foreign material in the hydrometeor as suggested by the homogeneous mixing assumption.
Due to sulfate production in the aqueous phase, all processed particles contain some sulfate
mass and therefore are attributed to the unactivated internally mixed/soluble modes. Adopting
lognormal size distributions for the newly formed aerosols, the dry median radius is calculated
following Hoose et al. (2008a) such that

\[
\tau_j = \sqrt[3]{\frac{3}{4\pi} \frac{V_j^{\text{tot}}}{N_j} \frac{1}{\exp\left(1.5 \ln^2 \sigma_j\right)}}, \quad j \in \{CD, IC, RD, SF, GR\}
\] (3.5)
$N_j$ denotes the number density of potentially released aerosol particles (equal to the number density of the corresponding hydrometeors) and $\sigma_j$ is the geometric standard deviation of the size distribution being fixed to the value of the corresponding unactivated aerosol mode. $V^\text{tot}_j$ represents the total in-hydrometeor volume of aerosol mass within the grid box as given by

$$V^\text{tot}_j = \sum_i \frac{M^X_j}{\rho^X_j}, \quad X \in \{\text{SU}, \text{BC}, \text{OC}, \text{SS}, \text{DU}\}. \quad (3.6)$$

The dry densities of the compounds are given by $\rho_{\text{SU}} = 1.841 \text{ g cm}^{-3}$, $\rho_{\text{BC}} = \rho_{\text{OC}} = 2.0 \text{ g cm}^{-3}$, $\rho_{\text{SS}} = 2.165 \text{ g cm}^{-3}$ and $\rho_{\text{DU}} = 2.65 \text{ g cm}^{-3}$ (Hoose et al., 2008a). The released aerosol particle is added to the unactivated soluble mixed mode with $\tau_m$ falling in its size range (Table 3.1).

### 3.3 Model setup

The present study focuses on the simulation of moist flow over two bell-shaped mountains in order to investigate the influence of processed aerosols on orographic cloud formation. The setup is based on the works of Muhlbauer and Lohmann (2008) and Muhlbauer et al. (2010). The two-dimensional computational domain comprises 500 horizontal grid points with a grid spacing of 2 km yielding a domain size of 1000 km. In the vertical, 50 levels follow hybrid height-based Gal–Chen coordinates from the surface to the top of the computational domain at 22 km. The vertical grid spacing varies between 10 m for the lowest model level and 1000 m for the highest level. The model time step is 10 s. At the bottom of the domain we impose a free-slip boundary condition. To minimize the reflection of gravity waves from the upper model boundary, a Rayleigh damping sponge layer fills the upper part of the computational domain beginning at an altitude of 11 km. The idealized topography consists of two identical bell-shaped mountains, each following the form given by Kirshbaum and Durran (2004):

$$h(x) = \begin{cases} \frac{h_0}{16} \left[ 1 + \cos \left( \pi \frac{x-x_i}{4a} \right) \right]^4, & |x - x_i| < 4a \\ 0, & |x - x_i| > 4a \end{cases} \quad (3.7)$$

with $h_0 = 800 \text{ m}$ the mountain peak height, $a = 20 \text{ km}$ the mountain half-width and $x_i$ the horizontal position of the mountain peak with $i \in \{1, 2\}$. The two mountain peaks are located at $x_1 = 600 \text{ km}$ and $x_2 = 800 \text{ km}$, implying a distance of 200 km between the two mountain peaks. In the present setup, aerosol particles may be processed by the cloud over the first mountain, and then advected towards the second mountain where the processed aerosol particles potentially influence further cloud formation.
3.3. Model setup

3.3.1 Dynamical initialization

The temperature and humidity profiles are initialized following the work of Muhlbauer and Lohmann (2008). The initial temperature profile is calculated analytically following Clark and Farley (1984) based on the sea-level pressure $p_{SL}$, sea-level temperature $T_{SL}$ and dry Brunt–Väisälä frequency $N_d$. The sea-level pressure is fixed to $p_{SL} = 1000$ hPa and the dry Brunt–Väisälä frequency is prescribed vertically constant with $N_d = 0.011$ s$^{-1}$ (Muhlbauer et al., 2010). The sea-level temperature varies between $T_{SL} = 280$ K and $T_{SL} = 270$ K to allow for the formation of warm-phase and mixed-phase clouds. The relative humidity profile is calculated using a modified Fermi function (Spichtinger, 2004) given by

$$RH(z) = a + \frac{b - a}{1 + \exp(-c(z - z_0))},$$

with the parameters $a = 0.90$, $b = 0.03$, $c = 0.0015$ m$^{-1}$ and $z_0 = 5000$ m. At the surface, the prescribed relative humidity profile has a value of $RH = 0.90$ which smoothly decreases with height to a value of $RH = 0.03$. The corresponding vertical profiles of temperature (red) and dew point temperature (blue) are shown in Fig. 3.2.

Solid lines indicate the cold profiles and dashed lines indicate the warm profiles. Following the setup of Muhlbauer et al. (2010) the horizontal wind speed $U$ is vertically constant with $15$ m s$^{-1}$ up to an altitude of $10$ km. It then increases linearly with height up to $40$ m s$^{-1}$ at the top of the model domain. The initial conditions as specified yield a dimensionless mountain height $N_d h_0/U = 0.59$. According to theoretical considerations of dry flow over topographies (e.g., Durran, 1990), one thus expects an unblocked regime with flow over the ridge and associated formation of gravity waves. In our simulations, however, the dynamics of the flow will be modified by latent heat release resulting from condensation. As we focus on microphysical effects only, the complexity of this aerosol–cloud interaction study is reduced by turning off the radiation scheme and the convection parameterization.

3.3.2 Aerosol initialization

The initial aerosol number concentrations are prescribed based on observations in order to simulate realistic atmospheric aerosol conditions (Fig. 3.3). The aerosol number concentrations are compiled from measurement data collected by a scanning mobility particle sizer (SMPS) during a field campaign in 1999 at the high Alpine research station Jungfraujoch (Weingartner et al., 1999). Throughout this study we use the average summertime aerosol size distribution.
Figure 3.2: Skew-T-log-P diagram of the atmospheric soundings for the idealized 2-D simulations showing the temperature (red) and the dew point temperature (blue) with a surface relative humidity of 90%. The warm sounding (dashed) with a surface temperature of 280 K is used for the simulation of a warm-phase cloud and the cold sounding (solid) with a surface temperature of 270 K is used for the simulation of a mixed-phase cloud.

as described by Muhlbauer and Lohmann (2008), which is representative of planetary boundary layer air in the Alpine region. During the summer months, convective processes and local circulations allow for the transport of boundary layer air including local anthropogenic emissions to the free troposphere (Weingartner et al., 1999). The initial aerosol composition is based on observations of aerosol mass spectrometry at the same location (see Fig. 11 in Cozic et al., 2008). Aerosol particles are internal mixtures of black carbon (BC), organic carbon (OC), sulfate (SU) and dust (DU). The first three compounds are, besides nitrates, frequently observed in central Switzerland (Hueglin et al., 2005). Dust can be occasionally observed at the Jungfraujoch during intense Saharan dust events (Cozic et al., 2008; Chou et al., 2011). The aerosol number and mass concentrations are set as constant with height based on the parameters for the aerosol size distributions given in Table 3.3.

Sulfate nucleation-mode particles are formed by gas-to-particle conversion from gas-phase sulfuric acid at low temperatures. Due to their small size, these particles cannot be measured by the SMPS. The vertical profiles of gaseous sulfate and the nucleation-mode aerosol particles are therefore initialized with averaged pseudo-soundings taken from the global circulation model ECHAM5-HAM (Stier et al., 2005) for the nearest Jungfraujoch grid point as described by Muhlbauer and Lohmann (2008).
3.3. Model setup

<table>
<thead>
<tr>
<th>Aerosol mode</th>
<th>$N_a$</th>
<th>$r$</th>
<th>$\sigma$</th>
<th>$M_a$</th>
<th>Mass fractional composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aitken</td>
<td>530 cm$^{-3}$</td>
<td>0.022 µm</td>
<td>2.13</td>
<td>0.27 µg m$^{-3}$</td>
<td>66% OC, 30% SU, 4% BC</td>
</tr>
<tr>
<td>Accumulation</td>
<td>260 cm$^{-3}$</td>
<td>0.070 µm</td>
<td>1.61</td>
<td>1.74 µg m$^{-3}$</td>
<td>60% OC, 28% SU, 9% DU, 3% BC</td>
</tr>
</tbody>
</table>

Table 3.3: Parameters of the initial aerosol size distributions (Muhlbauer and Lohmann, 2008) and mass fractional composition (Cozic et al., 2008) with $N_a$ the aerosol number density, $r$ the mode mean radius, $\sigma$ the standard deviation and $M_a$ the aerosol mass density of the lognormal distribution. Aerosol compounds include organic carbon (OC), black carbon (BC), sulfate (SU) and dust (DU).

Figure 3.3: Initial aerosol (a) number density and (b) mass density size distributions for all simulations.

To investigate the impact of aerosol processing and regeneration on the aerosol size distribution and further cloud formation, we vary the complexity of the representation of aerosol processes within the model (Table 3.4). The control simulations, CTL-w and CTL-c, employ the standard version of the aerosol module M7 without any scavenging processes (Muhlbauer and Lohmann, 2008, 2009) for the warm (w) and cold (c) cases, respectively. The simulations SCAV-w and SCAV-c include aerosol activation scavenging and below-cloud scavenging as described by Zubler et al. (2011a). In the simulations AP-w and AP-c we explicitly account for the new aerosol processing scheme including detailed aerosol activation, in-cloud and below-cloud scavenging processes, aerosol processing in clouds and aerosol regeneration upon hydrometeor evaporation and sublimation as described in the model section of this paper. To isolate the effect of aerosol regeneration, we additionally conduct simulations with the same parameterizations of the scavenging processes as in simulation AP-w and AP-c, but without aerosol regeneration (SCAV-ALL-w and SCAV-ALL-c).
Chapter 3. Microphysical processing of aerosol particles in orographic clouds

<table>
<thead>
<tr>
<th>Simulation</th>
<th>Temperature</th>
<th>Aerosol initialization</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CTL-w</td>
<td>280 K</td>
<td>all aerosols mixed</td>
<td>standard simulation (Muhlbauer and Lohmann, 2008, 2009) with aerosol activation and below-cloud scavenging (Zubler et al., 2011a)</td>
</tr>
<tr>
<td>SCAV-w</td>
<td>280 K</td>
<td>all aerosols mixed</td>
<td>with activation scavenging, the adapted below-cloud scavenging parameterization and in-cloud collision scavenging</td>
</tr>
<tr>
<td>SCAV-ALL-w</td>
<td>280 K</td>
<td>all aerosols mixed</td>
<td>with aerosol processing (including all scavenging processes and aerosol regeneration)</td>
</tr>
<tr>
<td>AP-w</td>
<td>280 K</td>
<td>all aerosols mixed</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Simulation</th>
<th>Temperature</th>
<th>Aerosol initialization</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CTL-c</td>
<td>270 K</td>
<td>50 % of DU is insoluble</td>
<td>standard simulation with aerosol activation and below-cloud scavenging</td>
</tr>
<tr>
<td>SCAV-c</td>
<td>270 K</td>
<td>50 % of DU is insoluble</td>
<td>with activation scavenging, the adapted below-cloud scavenging parameterization and in-cloud collision scavenging</td>
</tr>
<tr>
<td>SCAV-ALL-c</td>
<td>270 K</td>
<td>50 % of DU is insoluble</td>
<td>with aerosol processing</td>
</tr>
<tr>
<td>AP-c</td>
<td>270 K</td>
<td>50 % of DU is insoluble</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.4: Description of the conducted simulations.

3.4 Results

The following analyses are conducted after 10 h of simulation time, when a steady flow has been established and processed air masses from the first mountain pass over the second mountain. After 10 h, a hydrostatic mountain wave has developed with upstream regions of flow deceleration and downstream regions of flow acceleration. The upstream region is characterized by almost horizontal isentropes while the gravity wave generates foehn-like winds on the leeward side of the ridge.

3.4.1 Warm-phase cloud

This section focuses on the simulations initialized with the warm temperature profile with a surface temperature of 280 K. Similar dynamical conditions in all four simulations lead to the formation of warm-phase orographic clouds on the upslope side of the mountains with comparable liquid water mixing ratios (not shown). In the control simulation CTL-w, the cloud water mixing ratio has a maximum value of 0.97 g kg\(^{-1}\) over the mountain ridge (Fig. 3.4), whereas the rainwater mixing ratio does not exceed 0.01 g kg\(^{-1}\) (not shown). In the other simulations comparable values are produced at the first mountain. In a similar setup, Xue et al. (2010) obtained a maximum cloud water mixing ratio of more than 0.7 g kg\(^{-1}\) and a maximum rainwater mixing ratio between 0.01 g kg\(^{-1}\) for a dry and 0.12 g kg\(^{-1}\) for a wet environment. The generated clouds are almost entirely composed of cloud droplets, producing only a negligible amount of rain. Newly formed raindrops subsequently evaporate within the same time step as
no significant amount of precipitation remains in the atmosphere or falls to the surface.

**Figure 3.4:** Vertical cross section of the cloud water mixing ratio (QC) for the simulation CTL-w after 10h. Black lines indicate the potential temperature (κ) and red lines the temperature (K). *Only part of the computational domain is shown.*

The four simulations differ regarding the treatment of aerosol scavenging and regeneration processes which directly impacts the aerosol population. This becomes apparent in the number concentration of internally mixed/soluble aerosol particles larger than 35 nm $N_{>35\text{ nm}}$ (Fig. 3.5), which represents the number concentration of potential CCN in the model. In simulation CTL-w, where aerosol scavenging and regeneration processes are neglected, the aerosol number concentration $N_{>35\text{ nm}}$ is only affected by the dynamical flow over the mountain leading to a minor variation between 325 and 400 particles per cm$^3$. The impact of activation and below-cloud scavenging becomes apparent in simulation SCAV-w where $N_{>35\text{ nm}}$ is significantly reduced down to 200 particles per cm$^3$ in air masses which have passed through a cloud. Cloud droplet formation and collisions between aerosol particles and sedimenting hydrometeors reaching the surface efficiently scavenge aerosol particles in the region above the mountains. The corresponding air masses are then advected downstream so that reduced aerosol concentrations can be observed in the entire lower part of the model domain downstream of the first mountain. Simulation SCAV-ALL-w includes in-cloud collision scavenging and a more explicit treatment of below-cloud scavenging, as collisions with all sedimenting hydrometeors and not only with the ones reaching the surface are considered. In this simulation, $N_{>35\text{ nm}}$ shows a similar pattern. In-cloud collisions mainly involve Aitken-mode particles, which only represent a small subset of $N_{>35\text{ nm}}$. In simulation AP-w, some of the scavenged aerosol particles are replenished by aerosol regeneration upon evaporation in the downdraft region at the downslope side of the mountains. Since the prevailing wind in the simulations is unidirectional, most of the regenerated aerosol particles released upon evaporation in the first cloud are directly advected
downstream towards the second mountain where they may impact further cloud formation.

Figure 3.5: Vertical cross section of the number concentration of aerosol particles larger than 35 nm ($N_{>35\text{ nm}}$) which may potentially act as CCN in the model for simulation CTL-w, SCAV-w, SCAV-ALL-w and AP-w after 10 h. Black lines indicate the potential temperature ($\Theta$) and red lines the temperature ($T$). Only part of the computational domain is shown.

However, before being released upon evaporation, in-hydrometeor aerosol particles are subject to aerosol processing within the cloud. The transfer rates for in-hydrometeor aerosol mass illustrate the importance of the different microphysical processes for cloud processing of aerosol mass. The aerosol mass transfer rates after 10 h of simulation time for the cloud over the first mountain are shown in Fig. 3.6. Aerosol activation denotes the condensation of water vapor onto aerosol particles leading to the formation of cloud droplets. Aerosol activation and thus cloud droplet formation occur on the upslope side of the mountain between 60 and 20 km ahead of the mountain crest (540–580 km) where the cloud starts to form. Other processes which can potentially transfer aerosol mass from the unactivated aerosol modes into the in-hydrometeor modes are in-cloud impaction scavenging and below-cloud scavenging. However, in the present cloud, their contributions to the total in-hydrometeor aerosol mass remain negligible. A large part of in-droplet aerosol mass is released back to the unactivated aerosol modes upon cloud droplet evaporation. Most of the aerosol re-emission occurs behind the mountain crest between...
600 and 630 km on the downslope side of the mountain. The other microphysical processes generate insignificant transfer rates.

![Figure 3.6: Vertically integrated aerosol mass transfer rates per second of different processes within the microphysical scheme along the flow direction after 10 h in simulation AP-w over the first mountain (mountain peak represented by the triangle at 600 km). Only aerosol mass transfer rates larger than 0.2 µg m$^{-2}$ s$^{-1}$ are shown.](image)

The recycled aerosol particles are re-attributed to the free aerosol modes changing the size distribution and total number concentration of the interstitial or unactivated aerosol particles. Figure 3.7 depicts the vertical profile of $N_{>35 \text{ nm}}$ upstream (left panel) and downstream (right panel) of the first cloud. Upstream of the first mountain, the four simulations exhibit the same constant vertical profile of $N_{>35 \text{ nm}}$ with an average value of 360 particles per cm$^3$. After passage over the first mountain, the simulations show distinctly different vertical profiles. In simulation CTL-w the almost-constant vertical profile is conserved. The other simulations reveal a reduction in $N_{>35 \text{ nm}}$ from the surface up to an altitude of 3700 m. Simulations SCAV-w and SCAV-ALL-w include an almost-constant reduction to around 220 particles per cm$^3$ with considerable variations in the lower 1000 m. The strong peak at about 600 m altitude corresponds to new droplet formation between the two mountains. As the differences between these two simulations remain small, in-cloud scavenging and the adapted below-cloud scavenging parameterization accounting for all aerosol–hydrometeor collisions have only a negligible impact on $N_{>35 \text{ nm}}$ in the present setup. In simulation AP-w, the loss in $N_{>35 \text{ nm}}$ is partly replenished, maintaining a similar pattern in the vertical profile to the simulations SCAV-w and SCAV-ALL-w.

The total aerosol mass density also shows varying vertical profiles for the different simulations downstream of the first mountain (Fig. 3.8 right panel). In simulation CTL-w the vertical profile is conserved upstream and downstream of the mountain. Simulations SCAV-w and SCAV-ALL-w reveal similar vertical profiles with a reduction of the total aerosol mass density up to an altitude of 3700 m. The observed variations in the lower 1000 m due to new cloud formation between
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Figure 3.7: Vertical profile of the number concentration of aerosol particles larger than 35 nm ($N_{>35\, \text{nm}}$) after 10 h (a) upstream (averaged between 510 and 530 km) and (b) downstream (averaged between 680 and 720 km) of the first mountain for the warm cloud cases. Dotted horizontal lines indicate altitudes at which cumulative aerosol size distributions are compiled.

The mountains are also reflected in the profiles of the aerosol mass density. Simulation AP-w shows that most of the scavenged aerosol mass is replenished by evaporation throughout the column. In the lowest 1000 m the vertical profile follows the same pattern as in simulation SCAV-w and SCAV-ALL-w.

Figure 3.8: Vertical profile of the total aerosol mass density after 10 h (a) upstream (averaged between 510 and 530 km) and (b) downstream (averaged between 680 and 720 km) of the first mountain for the warm cloud cases. Dotted horizontal lines indicate altitudes at which cumulative aerosol size distributions are compiled.

The mean cumulative aerosol number and mass concentrations of the Aitken, accumulation
3.4. Results

and coarse modes downstream of the first orographic cloud at 300 and 2000 m altitude are depicted in Figs. 3.9 and 3.10. The size distributions are calculated from the simulated values for the number concentrations and dry radii of the different aerosol modes. To obtain representative values for the processed air masses after passage over the mountain, the variables are averaged horizontally between 680 and 720 km. We adopt the standard deviation from the initial measurements for the nucleation ($\sigma = 2.13$) and Aitken modes ($\sigma = 1.61$) and the prescribed value in COSMO-Model for the coarse mode ($\sigma = 2.0$). The number size distributions (Figs. 3.9a and 3.10a) are characterized by a bimodal shape of Aitken- and accumulation-mode particles. Differences between the initial aerosol number concentration and CTL-w are due to aerosol coagulation and sedimentation processes, as the corresponding air masses have been exposed to aerosol microphysical processes in the model domain for 10 h. After 10 h the bimodal shape of the aerosol number concentration becomes more pronounced and the number of aerosol particles in both modes is reduced. The aerosol mass distribution is dominated by accumulation-mode particles, as Aitken-mode particles are too small to contribute significantly to the total aerosol mass. A slight reduction of the aerosol mass distribution can be observed in CTL-w at 2000 m altitude (Fig. 3.10b). In simulation SCAV-w the scavenging processes within the orographic cloud impact the aerosol size distribution by reducing the number and mass of unactivated aerosol particles. Due to activation scavenging, $N_{>35 \text{ nm}}$ is removed from the atmosphere, reducing primarily the accumulation-mode number concentration and shifting the mean size of the Aitken-mode towards smaller sizes. The two aerosol modes appear clearly separated and the bimodal shape of the aerosol number concentration becomes more distinct. The scavenging processes also cause a decrease in the aerosol mass distribution. In simulation SCAV-ALL-w the adapted parameterizations for below-cloud scavenging and in-cloud collision scavenging between interstitial aerosol particles and cloud droplets induce an additional decrease in Aitken-mode particles compared to simulation SCAV-w. A sensitivity test revealed that this reduction is mainly due to in-cloud collision scavenging. No significant impact on the aerosol mass distribution can be observed as Aitken-mode particles account for only a negligible fraction of the aerosol mass due to their small size. In simulation AP-w, part of the processed aerosol particles is regenerated upon evaporation, increasing the number concentration and mass of aerosol particles compared to SCAV-ALL-w. The release of processed aerosol particles significantly increases the number concentration of the Aitken and accumulation modes along with a gain in the aerosol mass distribution compared to SCAV-ALL-w. Regenerated Aitken-mode particles have an increased size and mass compared to the background Aitken-mode particles. Therefore, the average radius of the combined Aitken mode is
increased shifting the lognormal size distribution towards larger sizes. The Aitken mode thus appears as a hump on the left-hand side of the accumulation-mode size distribution. At 2000 m altitude, the aerosol mass distribution reveals the formation of some large coarse-mode particles that impact the aerosol mass distribution, but not the aerosol number concentration.

Figure 3.9: Cumulative aerosol (a) number and (b) mass size distribution after cloud passage after 10 h for the warm cloud cases. The values are averaged horizontally between 680 and 720 km at 300 m altitude.

Figure 3.10: Cumulative aerosol (a) number and (b) mass size distribution after cloud passage after 10 h for the warm cloud cases. The values are averaged horizontally between 680 and 720 km at 2000 m altitude.

As aerosol particles act as CCN for cloud droplet formation, the changes in the aerosol size distributions impact cloud formation at the second mountain. The cloud droplet number concentration in simulation AP-w after 10 h over the second mountain and the difference in cloud droplet number concentration between simulations AP-w and SCAV-ALL-w are depicted in Fig. 3.11a and b. In simulation AP-w, significantly more cloud droplets are formed than in SCAV-ALL-w. As the liquid water content of the cloud is only marginally influenced by the modified aerosol
3.4. Results

concentrations, this translates in a larger number of droplets, which are smaller sized. Changes in the droplet size distribution potentially affect rain formation. However, as the present cloud is characterized as non-precipitating with no measurable rainwater content, a potential reduction of rainwater content and raindrop number concentration cannot be observed.

Figure 3.11: Number concentration of cloud droplets in (a) simulation AP-w and difference in cloud droplet number concentration between simulations (b) AP-w and SCAV-ALL-w, (c) AP-w and CTL-w and (d) AP-w and SCAV-w after 10 h over the second mountain. Black lines indicate the potential temperature (K) and red lines the temperature (K). Only part of the computational domain is shown.

The difference in cloud droplet number concentrations over the second mountain between simulations AP-w and CTL-w is shown in Fig. 3.11c. As already seen earlier in this paper, \( N_{>35 \text{ nm}} \) is reduced in AP-w compared to CTL-w. This results in a strong diminution of the cloud droplet number concentration. Figure 3.11d illustrates the difference in cloud droplet number concentrations over the second mountain between simulations AP-w and SCAV-w. The results are similar to Fig. 3.11b where the difference between simulations AP-w and SCAV-ALL-w is shown. Based on similar vertical profiles of \( N_{>35 \text{ nm}} \) in simulation SCAV-ALL-w and SCAV-w, comparable cloud droplet number concentrations are produced in both simulations. This result illustrates that in-cloud collision scavenging impacts the aerosol size distributions, but has only marginal influence on cloud droplet formation. Obviously, the lowest cloud droplet number
concentrations are observed in SCAV-w and SCAV-ALL-w. As no aerosol regeneration is taken into account, all newly formed clouds droplets at the second mountain activated from aerosol particles that have not been activated at the first mountain. Xue et al. (2010) observed a similar relationship between aerosol regeneration and cloud droplet number concentration.

**Sensitivity tests**

In order to evaluate the uncertainties of the presented results sensitivity studies are carried out. A first set of simulations is initialized with wintertime aerosol size distributions following Muhlbauer et al. (2010) and Xue et al. (2010, 2012) which represent a clean aerosol case. Accordingly, the number concentrations of the Aitken and accumulation modes are reduced to 310 and 40 cm$^{-3}$ respectively. The simulations generate precipitating warm-phase clouds with a maximum cloud liquid water mixing ratio of 0.76 g kg$^{-1}$ and a maximum rainwater mixing ratio of 0.10 g kg$^{-1}$ at the first mountain. As expected, the cloud water content is reduced and the rainwater content is increased in the clean simulations as compared to the more polluted warm case presented before. Aerosol mass transfer rates for the corresponding simulation with aerosol processing are depicted in Fig. 3.12. In this simulation, the aerosol mass transfer rates which are caused by activation scavenging and aerosol regeneration upon cloud droplet evaporation prevail. Aerosol mass transfer into raindrops caused by collisions and autoconversion, and the subsequent removal through precipitation or regeneration through evaporating raindrops play a minor role. In a cloud with enhanced rainwater content, these rates might increase.

![Figure 3.12: Vertically integrated aerosol mass transfer rates per second of different processes within the microphysical scheme along the flow direction after 10 h in the simulation with aerosol processing for the clean aerosol initialization over the first mountain (mountain peak represented by the triangle at 600 km). Only aerosol mass transfer rates larger than 0.02 µg m$^{-2}$ s$^{-1}$ are shown.](image)

The cumulative aerosol number and mass size distributions for the different clean case simu-
lations averaged between 680 and 720 km at an altitude of 300 m are displayed in Fig. 3.13. Aerosol scavenging in simulation SCAV and SCAV-ALL reduces the number of aerosol particles in the Aitken and accumulation mode, and it shifts the Aitken mode towards smaller radii. The impact of aerosol regeneration can be identified as a small increase in the number concentrations of both aerosol modes and a shift of the Aitken mode towards larger radii. These findings agree with the results of the polluted warm case.

![Figure 3.13: Cumulative aerosol (a) number and (b) mass size distribution after cloud passage after 10 h for the warm simulations with clean aerosol initialization. The values are averaged horizontally between 680 and 720 km at 300 m altitude.](image)

In the employed model configuration, the standard deviations of the log-normal aerosol size distributions are based on observations ($\sigma = 1.59$ for the nucleation, $\sigma = 2.13$ for the Aitken, $\sigma = 1.61$ for the accumulation, and $\sigma = 2.0$ for the coarse mode). To investigate the dependence of the results on the standard deviation, we conducted a set of simulations employing the M7 standard deviations which are 1.59 for the nucleation, Aitken and accumulation and 2.0 for the coarse mode. The standard deviations have fixed values in the model. Further sensitivity experiments include simulations with a reduced surface relative humidity of 85 and 80 %.

In order to evaluate the impact of aerosol processing and regeneration on the aerosol population and on the number concentration of possible CCN in the model, the aerosol population in a sample domain downstream of the first mountain between 680 and 720 km is analyzed. The results at 300 and 2000 m altitude are summarized in Table 3.5. $N_{\text{reg}}$ denotes the difference in the total aerosol particles number concentration averaged over the sample area between simulations AP and SCAV-ALL and thus represents the additional particles due to aerosol processing and regeneration. This number concentration is compared to the total aerosol number concentration $N_{\text{total}}$ (Aitken, accumulation and coarse mode) in the sample area in simulation AP. A similar approach is applied for CCN by comparing the difference in the CCN number con-
Chapter 3. Microphysical processing of aerosol particles in orographic clouds

Table 3.5: Ratios of the aerosol number concentration of “regenerated” particles ($N_{\text{reg}}$, difference between AP and SCAV-ALL), the total aerosol number concentration in simulation AP ($N_{\text{total}}$), the number concentration of “regenerated” particles potentially acting as CCN ($N_{\text{CCN, reg}}$, difference between AP and SCAV-ALL), the number concentration of CCN in simulation AP ($N_{\text{CCN, total}}$) and the number concentration of “missing particles due to scavenging” ($N_{\text{scav}}$, difference between SCAV-ALL and CTL) of the sensitivity studies of the warm case. The values are averaged horizontally between 680 and 720 km at 300 and 2000 m altitude. $\sigma^*$ denotes the simulations with the M7 standard deviations. The first two lines describe the analyzed warm case.

In order to evaluate how many of the scavenged aerosol particles are recycled by the cloud, the difference of the aerosol concentrations between simulation SCAV-ALL and CTL ($N_{\text{scav}}$) is calculated. $N_{\text{scav}}$ represents the missing particles due to scavenging processes. The relationship between $N_{\text{reg}}$ and $N_{\text{scav}}$ thus shows how many of the missing particles due to scavenging are replenished by regenerated particles. In the polluted experiments, this proportion ranges between 24 and 39% representing about one-quarter to one-third of the missing aerosol particles due to scavenging. In the clean experiments, it does not exceed 27%. These findings are lower than in the study of Xue et al. (2010), who found that regenerated particles replenish one-third to two-thirds of the missing particles.

The presented sensitivity experiments show that the impact of regenerated aerosol particles...
on the aerosol and CCN number concentrations depends on the cloud regime and the aerosol population. Variations in the relative humidity and the standard deviation are shown to have a smaller influence.

3.4.2 Mixed-phase cloud

We employ the cold temperature profile with a surface temperature of 270 K to generate mixed-phase orographic clouds over the two mountains. The ascent over the bell-shaped mountains results in the formation of liquid cloud droplets on the upslope side of the mountain. After 10 h, liquid droplets only persist at cloud base (Fig. 3.14). Advedted aloft, they may undergo heterogeneous freezing processes to form ice crystals which constitute the upper part of the cloud. Due to the difference in saturation vapor pressure over ice and over liquid, cloud droplets may also evaporate in the presence of ice crystals allowing for the enhanced condensation of water vapor on preexisting ice crystals (Wegener–Bergeron–Findeisen process). The cloud over the first mountain consists mainly of cloud ice and snow with little cloud liquid water. In this first cloud, collision processes between ice crystals lead to the aggregation of snowflakes which sediment to the surface. A large part of the cloud water is removed from the atmosphere by sedimenting snow, reducing the available water vapor in the atmosphere. Therefore, the environment downstream of the first cloud is drier than in front of the first cloud. The cloud over the second mountain developing in this drier environment has much less cloud ice and the snow formation processes are suppressed.

![Figure 3.14: Vertical cross section of the cloud water mixing ratio (QC), ice water mixing ratio (QI) and snow water mixing ratio (QS) in simulation CTL-c after 10 h. Black lines indicate the potential temperature (K) and red lines the temperature (K). Only part of the computational domain is shown.](image)

In mixed-phase clouds, a multitude of microphysical processes can take place which affect the aerosol population. In the present setup, the in-hydrometeor aerosol mass is concentrated in cloud droplets and snow, whereas the aerosol mass within ice crystals appears to be negli-
ble. Figure 3.15 illustrates which processes are important in terms of the transfer of aerosol mass into, out of and between the different hydrometeors. Aerosol mass is transferred into cloud droplets mostly by activation scavenging. When the cloud droplets reach higher altitudes some ice is formed heterogeneously via condensation/immersion freezing or contact freezing. Although these processes are essential for the formation of ice crystals in the model, their influence on the aerosol transfer remains negligible. In the presence of ice crystals, the liquid droplets rapidly evaporate and the water vapor condenses on the ice phase (Wegener–Bergeron–Findeisen process) when the vapor pressure is below water saturation. Therefore, cloud droplet activation and evaporation take place at the same horizontal location but at different altitudes. These two processes form a first aerosol processing cycle which acts via the liquid phase. The ice crystals grow and aggregate to form snowflakes which efficiently take up aerosol mass by riming with cloud droplets and below-cloud scavenging of interstitial aerosol particles. Sedimenting snowflakes remove aerosol mass from the atmosphere. Downstream of the cloud, some snowflakes sublimate releasing aerosol mass back to the interstitial mode (peak at 630 km). The second aerosol processing cycle therefore covers the interactions with snowflakes. A connection between the two cycles represents the riming process which transfers aerosol mass from cloud droplets to snowflakes. The other microphysical processes within the model are negligible for the transfer of aerosol mass in the present case study.

The described processes within the first cloud have an important influence on the vertical distribution of aerosol particles. Figure 3.16 shows the vertical profile of the total aerosol mass for the different simulations upstream (left panel) and downstream (right panel) of the first cloud. All simulations show the same vertical profile upstream of the first mountain. In simulation CTL-
3.4. Results

c, the vertical profile remains constant after passage over the mountain. Simulation SCAV-c includes a reduction of aerosol mass in the lower 3700 m where the cloud forms. This reduction is due to activation and below-cloud scavenging. The modified scavenging parameterizations in simulation SCAV-ALL-c generate a significantly larger diminution of the aerosol mass concentration in the lower part of the atmosphere. In simulation SCAV-ALL-c, below-cloud scavenging is treated differently, as all aerosol particles colliding with sedimenting rain or snow are removed from the unactivated aerosol modes, whereas in simulation SCAV-c only the fraction corresponding to the fraction of precipitation reaching the surface is removed. Therefore, below-cloud scavenging is far more efficient in simulation SCAV-ALL-c, as apparent in the amplified reduction of the vertical profile below 2000 m altitude. Additionally, SCAV-ALL-c accounts for in-cloud collision scavenging and scavenging of aerosol particles acting as contact IN. In simulation AP-c, evaporation processes lead to the release of additional aerosol particles, increasing the aerosol mass compared to SCAV-ALL-c. Above about 2200 m almost all of the scavenged aerosol mass is replenished by aerosol regeneration upon evaporation.

Figure 3.16: Vertical profile of the total aerosol mass density after 10 h (a) upstream (averaged between 510 and 530 km) and (b) downstream (averaged between 680 and 720 km) of the first mountain for the cold cloud cases. Dotted horizontal lines indicate altitudes at which cumulative aerosol size distributions are compiled.

The cumulative aerosol number and mass size distributions of the Aitken, accumulation and coarse modes downstream of the first mountain at 300 and 2000 m altitude are depicted in Figs. 3.17 and 3.18.

The values are averaged horizontally between 680 and 720 km. Aerosol microphysical processes like coagulation and sedimentation lead to a reduction in the aerosol number and mass...
size distribution after 10 h in simulation CTL-c compared to the initial aerosol size distributions. At both altitudes the bimodal shape of the aerosol number size distribution becomes more distinct while both Aitken and accumulation modes are reduced. The aerosol mass size distribution determined by the accumulation mode is slightly reduced due to sedimentation. Scavenging processes in simulation SCAV-c lead to a considerable reduction in aerosol number and mass. The influence on the shape of the aerosol number size distribution becomes particularly apparent at 300 m altitude, where Aitken and accumulation modes are clearly separated by a large gap. At this low altitude, the aerosol mass size distribution is significantly reduced. The reduction in aerosol mass and number at 300 m altitude is even more pronounced in simulation SCAV-ALL-c where the modified below-cloud scavenging parameterization, in-cloud collision scavenging and scavenging of contact IN remove the major part of unactivated aerosol particles from the atmosphere. A comparison with the non-precipitating warm-phase cloud suggests that an important part of the scavenging is due to the more detailed below-cloud scavenging by
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Sedimenting snow particles. At an altitude of 2000 m, below-cloud scavenging and activation scavenging become less important. The differences between AP and SCAV-ALL illustrate the impact of regenerated aerosol particles on the background aerosol size distributions. Aerosol regeneration upon droplet evaporation and snow sublimation in simulation AP-c leads to an increase in aerosol number and mass compared to simulation SCAV-ALL-c. The aerosol number size distribution reveals that large Aitken- and accumulation-mode particles are released to the atmosphere. Newly formed Aitken-mode particles have an increased size and mass as compared to the background Aitken-mode particles. The log-normal distribution of the Aitken mode is shifted towards larger size and appears as a bump on the left-hand side of the accumulation-mode size distribution. At 300 m altitude few coarse-mode particles are formed which appear in the aerosol mass distribution.

As aerosol particles serve as CCN and/or IN, changes in the aerosol number and mass size distribution may impact the number concentration of cloud droplets and ice crystals as shown in Fig. 3.19. The cloud droplet number and ice crystal number concentrations over the second mountain in simulation AP-c are displayed in Fig. 3.19a and b, differences between simulations AP-c and SCAV-ALL-c are shown in Fig. 3.19c and d. The released aerosol particles in AP-c lead to an increase in cloud droplet number concentration as compared to SCAV-ALL-c. The ice crystal number concentration reveals a comparable increase as more cloud droplets and also more aerosol particles are available for heterogeneous freezing processes. However, these changes in the number concentrations do not impact the cloud liquid and ice water content noticeably.

Sensitivity tests

Several sensitivity test were carried out in order to evaluate the uncertainties of the results. We conducted simulations with a lowered surface relative humidity to 85 and 80 %, with altered standard deviations, and with a clean (winter) aerosol initialization. The analysis focuses on the averaged aerosol population downstream of the first mountain between 680 and 720 km. The results at an altitude of 300 and 2000 m are summarized in Table 3.6. The difference between simulations AP and SCAV-ALL (\(N_{\text{reg}}\)) is compared to the total aerosol number concentration of the Aitken, accumulation and coarse modes (\(N_{\text{total}}\)) in simulation AP. Analogously, the difference in the CCN number concentration between simulation AP and SCAV-ALL (\(N_{\text{CCN}}^{\text{reg}}\)) is related to the CCN number concentration (\(N_{\text{total}}^{\text{CCN}}\)) in simulation AP. Regenerated aerosol particles constitute between 18 and 28 % of the total aerosol particles and between 38 and 57 % of the CCN particles in polluted simulations. Similar to the warm case, the clean sensitivity experiments of the cold case reveal a negligible contribution of regenerated particles to the total
aerosol concentration ($< 5 \%$), but constitute a considerably part of the CCN number concentration (26–55 \%). Further, we investigate how many of the missing particles due to scavenging are replenished by regenerated particles. $N_{\text{reg}}$ is compared to the difference of the aerosol concentrations between simulation SCAV-ALL and CTL ($N_{\text{scav}}$). In all sensitivity experiments at 300 m altitude, regenerated particles replenish only less than 10 \% of the missing particles, supporting the assumption that most of the scavenged particles are removed from the atmosphere by precipitation. At 2000 m altitude, a replenishment of up to 57 \% can be observed.

The results show that the cloud and the aerosol regime play an important role for aerosol regeneration and its impact on the background aerosol population. For precipitating clouds, the analyzed altitude is a determining factor.
### 3.5 Summary and conclusions

The effects of warm-phase and mixed-phase orographic clouds on the aerosol population have been evaluated by simulating orographic cloud formation over a 2-D double-bell-shaped topography with the regional weather forecast and climate model COSMO-Model. An explicit treatment of in-hydrometeor aerosol mass allowing for the consideration of aerosol processing in clouds has been implemented in the model. Aerosol scavenging processes and aerosol processing in clouds including aerosol regeneration upon evaporation or sublimation affect the aerosol population greatly. In this paper, different aerosol cycles have been identified. In the simulated non-precipitating warm-phase cloud, aerosol mass is incorporated in cloud droplets by activation scavenging and released back to the atmosphere upon cloud droplet evaporation.

In the simulated precipitating mixed-phase cloud, a significant amount of aerosol mass can be found in cloud droplets and snowflakes, but less in ice crystals. Activation and below-cloud scavenging efficiently transfer aerosol mass into the cloud droplets and snowflakes, while ice crystals are formed heterogeneously from a few cloud droplets allowing for only a limited transfer of aerosol mass. In the mixed-phase clouds, two aerosol cycles were identified. A first cycle includes activation scavenging and cloud droplet evaporation due to the WBF process. A second cycle includes the interactions with snowflakes and is connected to the first cycle via the riming process which transfers aerosol from cloud droplets to snowflakes. As aerosol parti-

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Table 3.6: Ratios of the aerosol number concentration of “regenerated” particles \((N_{\text{reg}}, \text{difference between AP and SCAV-ALL})\), the total aerosol number concentration in simulation AP \((N_{\text{total}})\), the number concentration of “regenerated” particles potentially acting as CCN \((N_{\text{CCN}}^{\text{reg}}, \text{difference between AP and SCAV-ALL})\), the number concentration of CCN in simulation AP \((N_{\text{CCN}}^{\text{total}})\) and the number concentration of “missing particles due to scavenging” \((N_{\text{scav}}, \text{difference between SCAV-ALL and CTL})\) of the sensitivity studies of the cold case. The values are averaged horizontally between 680 and 720 km at 300 and 2000 m altitude. \(\sigma^*\) denotes the simulations with the M7 standard deviations. The first two lines describe the analyzed cold case.
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cles are transported together with the sedimenting hydrometeors, a vertical redistribution of the aerosol number and mass concentration occurs with aerosol mass being transported towards lower altitudes or even being removed from the atmosphere. Precipitating snow in the lower atmosphere very efficiently removes aerosol number and mass from the atmosphere.

The present study shows that the different scavenging parameterizations and the consideration of aerosol regeneration modify the total aerosol number and mass and also the shape of the aerosol size distributions. Accounting for aerosol processing in clouds, the scavenged aerosol particles are partly replenished by the release of aerosol particles upon hydrometeor evaporation/sublimation. The released aerosol particles mainly add to the Aitken and accumulation mode. Depending on the simulated cloud, the formation of coarse-mode aerosol particles can be observed at specific altitudes. The newly generated large coarse-mode particles are possible candidates for giant cloud condensation nuclei (GCCN) which may lead to rapid rain formation in warm-phase clouds (Saleeby et al., 2009; Posselt and Lohmann, 2008). This in turn would impact the precipitation rate and the longevity of the cloud. However, the current model does not include any specific treatment of GCCN which would allow one to study the impact of these particles in more detail. The release of internally mixed aerosol particles in the accumulation and coarse modes increases the number concentration of possible CCN in the model impacting further cloud formation. The simulations show that aerosol regeneration increases the cloud droplet number concentration at the second mountain with possible implications for the ice crystal number concentration. In comparison to the standard model version, the aerosol activation and below-cloud scavenging processes revealed to be most important reducing the aerosol number concentration and in turn the cloud droplet number concentration at the second mountain. An influence on the ice crystal number concentration is observable via the changes in cloud droplet number concentration. In-cloud collision scavenging mainly affects the Aitken mode without any important implications for cloud droplet formation in warm-phase clouds. Due to collision processes, insoluble aerosol particles, which potentially may act as ice nuclei in the contact mode, are removed from the free atmosphere limiting the efficiency of contact freezing. However, in the present mixed-phase clouds this effect remains negligible.

Changes in the cloud droplet and ice crystal number concentration potentially affect the formation of precipitation in the model. However, in the present setup of non-precipitating warm-phase clouds, the increase in cloud droplet number concentration does not imply any important changes for the rain formation over the second mountain. In a precipitating cloud however, the formation of a greater number but smaller-sized cloud droplets may impede rain formation, as autoconversion and collision efficiencies may be reduced. The influence of the variations in
3.5. Summary and conclusions

cloud droplet and ice crystal number concentration on the ice water content in the simulated mixed-phase clouds remains negligible. Most of the ice is formed via the Wegener–Bergeron–Findeisen process which effectively adds water vapor to the ice crystals. Due to the high efficiency of this process in the present setup, the number of preexisting ice particles appears to be non-relevant for the total ice water content.

The present study focuses on the influence of aerosol processing in warm-phase and mixed-phase clouds on the aerosol population over two idealized 2-D bell-shaped mountains. A subsequent study should investigate the implications of the modified aerosol spectrum on further cloud formation in a 3-D regional study. Special emphasis should be placed on precipitation formation and the governing cloud microphysical processes.

Acknowledgements

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Edited by: C. Hoose
Chapter 4

Aerosol processing in Arctic springtime low-level clouds

4.1 Introduction

Arctic average surface temperatures have risen at almost twice the global rate during the past century (Boucher et al., 2013; McBean et al., 2005). A number of negative and positive climate feedbacks contribute to this unusually high sensitivity to climate change. Due to a unique feedback between aerosol forcing and springtime climate (Flanner et al., 2009) the spring season is of particular interest to Arctic climate change studies. From late winter to early spring, surprisingly high aerosol concentrations (Arctic haze) are observed in the Arctic atmosphere. Shaw (1982) first associated Arctic haze with polluted air masses from the industrialized South which are transported to the Arctic and trapped during the winter by the strong temperature inversions over the Arctic. In this cold, dry and stable system, the removal of particles and gases from the atmosphere is considerably slowed down leading to an accumulation of particles and gases in the Arctic atmosphere (Shaw, 1995). Strong seasonal cycles of aerosol concentrations with highest concentrations during winter were reported from Point Barrow in Alaska (Quinn et al., 2002), Alert in Canada (Gong et al., 2010) and Zeppelin Mountain on Svalbard (Eleftheriadis et al., 2009). Observations of cloud condensation nuclei (Leaitch et al., 1984; Yum and Hudson, 2001) and ice nuclei (Rogers et al., 2001; Prenni et al., 2009) concentrations reveal comparable seasonal patterns with increased numbers during winter and early spring compared to summer or early autumn. These variation potentially produce fluctuations in the cloud droplet and ice crystal number concentrations. The spring season between March and May represents a transitional period as the haze becomes less abundant and clouds more frequent and thick (McFarquhar et al., 2010). Curry et al. (1996) describe possible Arctic climate feedback processes
underscoring the importance of clouds to better understand radiation-climate feedbacks. Accordingly, the role of clouds and the associated feedbacks represent a key issue for simulating Arctic climate change. Clouds, including low-level mixed-phase clouds, constitute a prevalent feature of the Arctic atmosphere. In-situ observations of the Surface Heat Budget of the Arctic Ocean (SHEBA) experiment reveal a cloud occurrence of 85 % during the one year experiment (Intrieri et al., 2002). Likewise, cloud observations of one decade from 6 Atmospheric research stations in the Arctic showed a cloud occurrence of 58-93 % (Shupe et al., 2011) and satellite observations of the year 2007 and 2010 indicate an annual average cloud cover between 63.3 % and 78.4 % (Chan and Comiso, 2012).

Liquid and mixed-phase clouds are observed in the Arctic throughout the year, within a high range of temperatures and altitudes (Intrieri et al., 2002). 59 % of the clouds observed during the SHEBA campaign were identified as mixed-phase clouds, with a prevalence of this cloud type during the spring and autumn transition seasons (Shupe et al., 2006). Most mixed-phase clouds can be associated with temperatures between $-25^\circ$C and $-5^\circ$C (Shupe et al., 2006). They occur in single and multiple layers (Verlinde et al., 2007) covering large swaths throughout the year (Shupe et al., 2011) and can possibly persist for several hours to several days at a time (Shupe et al., 2006, 2011). Due to their horizontal extent and longevity, as well as their microphysical properties (Lubin and Vogelmann, 2011), mixed-phase clouds play an important role in the Arctic radiation budget.

Mixed-phase clouds consist of a mixture of supercooled droplets and ice particles. Due to the difference in vapor pressure over liquid water and ice, the mixture is unstable, leading to the growth of ice particles by vapor deposition at the expense of liquid water. This mechanism, known as the Wegener-Bergeron-Findeisen (WBF) process, can lead to a rapid glaciation of the cloud within a few hours or less. Considering the instability of liquid ice mixtures, the observed persistence of Arctic mixed-phase clouds is unexpected. The coexistence of liquid and ice can be explained by a balance between cloud droplet condensational growth, ice crystal depositional growth and ice removal by sedimentation. A conceptual model of Arctic mixed-phase clouds can be found in Shupe et al. (2008) and Morrison et al. (2012). Accordingly, turbulence and upward motions are assumed to play a key role in the maintenance of cloud liquid, as the air in moist updrafts can become supersaturated with respect to liquid water allowing for the formation of cloud droplets. Radiative cooling at cloud top is assumed to be a major driver for turbulence and updrafts, whereas the moisture supply can be assured by large-scale advection.

In spite of the importance of adequate simulation of Arctic clouds for the investigation of ques-
tions regarding Arctic climate change, the representation of Arctic mixed-phase clouds in climate models remains poor. Indeed, climate simulations in the Arctic reveal to be very sensitive to assumed cloud properties. To resolve differences between observed and modeled climate change, the complex interactions between clouds, aerosols and other components of the climate system must be better understood (McFarquhar et al., 2010).

Arctic mixed-phase clouds develop under temperature regimes warmer than the temperature for homogeneous freezing of \(-38\, ^\circ\text{C}\), indicating the importance of heterogeneous freezing processes in these clouds (Morrison et al., 2012). At this, aerosol particles play a crucial role, as they can act as cloud condensation nuclei (CCN) for the formation of cloud droplets or ice nuclei (IN) for heterogeneous ice nucleation. By influencing the number concentration and size distribution of supercooled liquid droplets at cloud top, aerosol particles impact the radiative properties of the cloud. Observations indicate that the increase in infrared emissivity of springtime Arctic clouds due to enhanced aerosol concentrations is climatologically significant (Lubin and Vogelmann, 2006). Changes in these properties can also have an effect on the persistence of mixed-phase clouds. Some aerosols also act as IN, although their concentrations is much lower than the CCN concentration. In mixed-phase cloud regimes, the presence of IN is essential for the formation of the first ice crystals. Additional ice crystals may be produced by secondary processes e.g. by Hallet-Mossop ice multiplication (Hallett and Mossop, 1974).

Modeling studies have shown that the balance between liquid and ice is sensitive to the ice crystal number concentration, and thus the concentration of heterogeneous ice nuclei. However, the correct simulation of ice crystal number concentrations based on observed IN concentrations remains challenging. Some recent studies showed that the observed IN concentrations are insufficient to explain the observed ice crystal number concentrations (e.g. Fridlind et al., 2007; Morrison et al., 2008; Fridlind et al., 2012) suggesting the existence of additional IN or ice crystal formation mechanisms. However, some studies also find agreement between observed and modeled ice number concentrations (e.g. Avramov et al., 2011).

Prenni et al. (2007) concluded from their modeling study that models must correctly represent not only the ice crystal number concentrations, but also cloud processing and removal of IN through precipitation, in order to capture the radiatively important liquid phase in Arctic clouds. When treated prognostically, IN concentrations are rapidly reduced due to ice nucleation. Additional IN sources help to maintain prognostic IN concentrations in the model. One approach consists of accounting for the recycling of IN from residuals of evaporating drops or sublimating ice crystals. Modeling studies reveal that accounting for this improves the representation of Arctic mixed-phase clouds (Fridlind et al., 2007) and can dramatically contribute to ice forma-
These results highlight the need for a most accurate representation of IN and CCN concentrations, and thus the aerosol size distribution and composition, in order to adequately simulate Arctic mixed-phase clouds. We take these findings as motivation for our work. Cloud-aerosol interactions not only impact cloud properties, but also modify the aerosol size distribution, mixing state and chemical composition. Due to activation into cloud droplets, heterogeneous ice nucleation and collision-coalescence processes aerosol particles are incorporated into hydrometeors. Processes like autoconversion, accretion, aggregation, freezing, melting, riming and self-collection transfer aerosol mass between the different hydrometeor classes. Aqueous phase chemistry within droplets can lead to the formation of sulfate mass. Finally, wet deposition, sedimentation and scavenging processes lead to a removal of aerosol mass from the atmosphere. However, a substantial part of hydrometeors evaporates/sublimes releasing a newly formed aerosol particle to the atmosphere with a different size, composition and mixing state as compared to the original one. Globally averaged, aerosol particles are cycled three times in a cloud before being removed from the atmosphere (Pruppacher and Jaenicke, 1995), including cycling in both stratiform and convective clouds. Global model simulations of aerosol cycling in stratiform clouds only showed an average of 0.5 cycles (Hoose et al., 2008b).

The presented study investigates the impact of aerosol processing in clouds on the simulated cloud properties for Arctic low-level liquid and mixed-phase clouds. The simulations are based on an extended model version including an explicit treatment of cloud-borne aerosol particles following the approach by Hoose et al. (2008a), in order to account for cloud cycling of aerosols by tracking the particles even when scavenged into hydrometeors.

After a brief model description (section 4.2), we provide the details of the simulation setup (section 4.3) and the field observations (section 4.4). In section 4.5, the results of the model simulations are presented and compared to measurements. We end with a summary and discussion of our findings in section 4.6.

4.2 Model description

In the present study, we apply the non-hydrostatic, fully compressible, limited-area mesoscale atmospheric prediction model of the COnsortium for Small-scale MOdeling (COSMO, http://www.cosmo-model.org). The model, originally developed by the German weather service
4.2. Model description

(DWD), is currently advanced and applied within the COSMO consortium. The simulations for the present study are performed using the climate version of the model known as CCLM (http://www.clm-community.eu).

The model is based on the primitive hydro-thermodynamical equations formulated on a rotated Arakawa C-grid (Doms and Schättler, 2002; Steppeler et al., 2003). For time integration, a split-explicit third order Runge-Kutta scheme is applied in combination with a 5th order upstream horizontal advection scheme. The aerosol and moisture variables are advected by the second order Bott scheme (Bott, 1989).

The employed model version includes a two moment bulk cloud microphysics scheme (Seifert and Beheng, 2006) which is coupled to the two-moment aerosol module M7 (Vignati et al., 2004; Muhlbauer and Lohmann, 2008, 2009; Zubler et al., 2011a; Pousse-Nottelmann et al., 2015). The cloud microphysical scheme includes prognostic equations for the mass and number densities of five different hydrometeor types: cloud droplets, rain drops, ice crystals, snow flakes and graupel. In warm-phase clouds, it accounts for cloud droplet activation, condensational growth and evaporation of cloud droplets, autoconversion and accretion, selfcollection, evaporation of rain and collisional break-up of large rain drops. Parameterized microphysical processes including the ice-phase are homogeneous and heterogeneous freezing, diffusional growth of ice crystals, aggregation, self-collection, riming, melting and sublimation as described by Seifert and Beheng (2006). The scheme also considers related secondary processes e.g. Hallet-Mossop ice multiplication (Hallett and Mossop, 1974), partial conversion of riming cloud ice to graupel, riming splintering and enhanced melting.

The M7 aerosol module predicts the evolution of mass and number densities of seven different aerosol modes which are represented by lognormal size distributions. The scheme distinguishes between four internally mixed, soluble modes (nucleation, Aitken, accumulation, and coarse mode) and three insoluble modes (Aitken, accumulation, and coarse mode). Five different aerosol compounds are accounted for, including sulfate (SU), carbonaceous aerosols (black carbon BC, organic carbon OC), sea salt (SS) and mineral dust (DU). At this, the insoluble modes contain externally mixed freshly formed black carbon and dust. Due to sulfate coating or coagulation, insoluble aerosols can be transferred to the mixed modes.

In the standard version of the model as described by Muhlbauer and Lohmann (2008, 2009), the aerosol modes contain both free unactivated and activated in-hydrometeor aerosol particles. The aerosol particles may act as cloud condensation nuclei or ice nuclei, but are not affected by cloud processes themselves. A second version of the model accounts for different aerosol
scavenging processes including activation scavenging and in-cloud and below-cloud collision scavenging (Zubler et al., 2011a; Pousse-Nottelmann et al., 2015). The model has recently been extended by an explicit treatment of in-hydrometeor aerosol particles (Pousse-Nottelmann et al., 2015). In this new model version, only unactivated aerosol particles are assigned to the M7 module, whereas activated in-hydrometeor particles are attributed to five new aerosol modes corresponding to the five hydrometeor classes. This distinction allows for the simulation of cloud cycling of aerosol particles. Due to activation or collision processes aerosol particles are incorporated into hydrometeors where they are subject to cloud microphysical processes. Upon evaporation / sublimation, the aerosol mass is released back to the atmosphere forming a newly generated aerosol particle.

### 4.3 Experimental setup

The present study focuses on the evaluation of the impact of aerosol processing in clouds on Arctic low-level liquid and mixed-phase clouds. Therefore, the model results are compared to in-situ observations acquired during the Indirect and Semi-Direct Aerosol Campaign (ISDAC) which took place in 2008 in Alaska.

The simulations for the present study are based on a one-way nesting approach. The model system employs the COSMO model for both the coarse and the fine resolution simulations with model domains centered over Barrow, Alaska ($71^\circ$N, $156^\circ$W). The coarse simulations are operated at a horizontal resolution of $0.11^\circ \times 0.11^\circ$ (approx. 12 km) on a rotated longitude-latitude grid. The model domain comprises 60 vertical levels, characterized by an increasing vertical layer thickness with altitude up to the top of the model atmosphere at approximatively 22 km. The model domain (150 x 175 grid points) covers the territory of the mainland of Alaska and the easternmost parts of Russia (Fig.4.1, left panel). The model time step is 90 s. At the lateral boundaries, the meteorological fields are driven by 6 hourly ECMWF ERA-Interim reanalysis data.

For the aerosol fields, we consider a simulation of the global circulation model ECHAM5-HAM (Stier et al., 2005). However, Bourgeois and Bey (2011) showed that the standard ECHAM model underestimates the long-range transport of aerosol particles from the mid-latitudes to the Arctic region leading to an underrepresentation of aerosol population in the Arctic. As we aim to conduct case studies with representative aerosol loadings we decided to prescribe the initial and lateral boundary conditions for aerosol concentrations based on observations. During the ISDAC campaign, aerosol particles larger than $0.12 \mu m$ were measured allowing for a specifi-
4.3. Experimental setup

Figure 4.1: Coarse and fine model domain with grid cell orography [m]. Left panel: Coarse model domain. The white rectangle indicates the boundaries of the fine model domain. Right panel: Fine model domain. White solid lines display the flight paths for the two case studies Case A and Case B, the white dashed rectangles represent the corresponding model sample areas.

cation of the accumulation and coarse mode. The aerosol number and mass concentrations were prescribed as vertically constant for both modes. Nucleation and Aitken mode remain based on the T106 ECHAM5-HAM global simulation. The applied lognormal fitting parameters for the aerosol accumulation and coarse mode (Table 4.1) were calculated by Earle et al. (2011) for measured aerosol size distributions during the ISDAC field campaign. The parameters are based on a compilation of below-cloud PCASP-100 (about 0.12 - 3 µm) and FSSP-300 aerosol measurements distinguishing between a polluted (Figure 4.2 a and b) and a clean case (Figure 4.2 c and d). A detailed description of the compilation of the measurements, measurement errors and implication for cloud droplet activation can be found in Earle et al. (2011). The mass fractional composition for accumulation and coarse mode is based on the aerosol composition used for the sensitivity tests in Liu et al. (2011) (Figure 4.3).

<table>
<thead>
<tr>
<th>regime</th>
<th>mode</th>
<th>$N_{tot}$ (cm$^{-3}$)</th>
<th>$r_m$ (µm)</th>
<th>$\sigma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>polluted</td>
<td>Accumulation</td>
<td>739</td>
<td>0.11</td>
<td>1.55</td>
</tr>
<tr>
<td></td>
<td>Coarse</td>
<td>8</td>
<td>0.29</td>
<td>2.16</td>
</tr>
<tr>
<td>clean</td>
<td>Accumulation</td>
<td>200</td>
<td>0.10</td>
<td>1.47</td>
</tr>
<tr>
<td></td>
<td>Coarse</td>
<td>10</td>
<td>0.36</td>
<td>2.44</td>
</tr>
</tbody>
</table>

Table 4.1: Table of the clean and polluted aerosol initial and boundary concentration for the coarse simulation. $N_{tot}$ is the total aerosol number concentration, $r_m$ is mode number mean radius (Earle et al., 2011).
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Figure 4.2: Aerosol number and mass size distribution of the accumulation (Acc.) and coarse mode for the polluted case (a and b) and the clean case (c and d) based on the lognormal fitting parameters from Earle et al. (2011).

Figure 4.3: Initial aerosol mass composition for the accumulation and the coarse mode based on Liu et al. (2011).

Dust and DMS aerosol emissions are based on the ECHAM5-HAM simulation, and BC, OC and SU emissions are taken from the Representative Concentration Pathways (RCP) aerosol emission scenario RCP 6.0 (Fujino et al., 2006; Hijioka et al., 2008; Vuuren et al., 2011). Sea salt emissions are calculated online. In the coarse simulation, we account for aerosol sedimentation and dry deposition, but do not explicitly consider aerosol processing in clouds.
4.4 Observations

The coarse simulation serves as initial and boundary data for the meteorological, cloud and aerosol fields of the fine simulations conducted at a horizontal resolution of 0.02° x 0.02° (approx. 2 km). The fine model domain includes 300 x 200 grid points, encompassing the northern part of Alaska and a part of the Arctic ocean (Fig.4.1, right panel). The 60 vertical levels correspond to the vertical levels of the coarse simulation. High resolution simulations are conducted with a time step of 10 s. Aerosol emissions are treated analogously to the coarse simulation.

We conduct two case studies to evaluate the model performance regarding the simulation of Arctic low-level liquid and mixed-phase clouds and to investigate the impact of aerosol processing in clouds on the aerosol and cloud properties. Throughout the study, we focus on the vertical structure of the cloud and aerosol properties. The simulations are started at 0 UTC the day before the observations allowing for a spinup time of 24 h.

We apply three different model versions of different complexity for the representation of aerosol-cloud interactions. In the standard version (STD), the aerosol population is not affected by cloud microphysical processes (Muhlbauer and Lohmann, 2008, 2009). A second model version includes activation, in-cloud and below-cloud collision scavenging (SCAV) as described by Zubler et al. (2011a) and Pousse-Nottelmann et al. (2015). In version AP the scavenging processes are completed by a detailed treatment of in-hydrometeor aerosol mass allowing for the simulation of aerosol cycling in cloud including aerosol regeneration upon hydrometeor evaporation / sublimation (Pousse-Nottelmann et al., 2015).

For the mixed-phase cloud case, we conduct sensitivity tests regarding the properties of the potential ice nuclei. At this, we account for different mixing states of the dust aerosol particles and consider bacteria as potential ice nuclei.

4.4 Observations

The Indirect and Semi-Direct Aerosol Campaign (ISDAC) campaign conducted in April 2008 in the vicinity of Barrow, Alaska allowed to collect a detailed set of field observations of spring-time Arctic clouds. The principal aim of the campaign was to advance our understanding of how changes in the composition and concentration of aerosols influence cloud microphysical and radiative properties in the Arctic (McFarquhar et al., 2010). For this purpose, the National research Council (NCR) of Canada Convair-580 aircraft equipped with cloud and aerosol instruments collected cloud and aerosol data on 12 different days under varying aerosol, surface
and meteorological conditions. Additional data was provided by radar and ground-based remote sensing observations. In the following, we briefly introduce the measurements employed in the present study. For a detailed description of the field campaign and the measurements that were carried out we refer to McFarquhar et al. (2010).

During the campaign, a Passive Cavity Aerosol Spectrometer Probe (PCASP-100X) measured ambient aerosol concentrations and size distributions of particles with diameters between 0.12 and 3 \( \mu \text{m} \). In clear air, the instrument sampled the total aerosol population, while within the cloud, only interstitial aerosol particles were measured. The instrument works by collecting scattered light from particles passing through the instrument's optics. After entering the instrument via a heated diffuser cone, the air sample passes through the object plane encountering a focused laser beam. When passing through the laser beam, the particles inside the air sample scatter light, which is measured by a photodetector and compared to a reference photodetector. Based on the Mie scattering theory the particle size is calculated and classified into one of the fifteen size bins from 120 \( \text{nm} \) to 3000 \( \text{nm} \). Uncertainties in the concentration measurements are related to flow measurement and estimated to be about 5 %. The uncertainty in particle sizing is related to the calibration of the instrument and estimated as the width of each size bin.

To distinguish between clear sky and cloudy observational data, we employ compiled cloud data indicating the phase of the cloud. The provided data was determined by a phase identification algorithm based on previously defined algorithms by McFarquhar and Cober (2004) and McFarquhar et al. (2007).

Cloud particles were measured by different instruments during the campaign. These observation were compiled by R. Jackson and G. M. McFarquhar to generate data for liquid water content (LWC), ice water content (IWC) and cloud droplet and ice crystal number concentration. The computation of the liquid water content is based on measurements of the cloud droplet number concentration and cloud droplet diameter. The ice water content is calculated using an automated habit identification scheme applied to CPI imagery that sorts crystals into habit classes based on their morphological characteristics. Details on the techniques can be found in Jackson (2011).

4.5 Comparison to observations

In this section we compare the model results to aircraft observations for two different case studies. We investigate time periods of about 2.5 hours averaging measurements and model results over the same period of time. By flying back and forth on the same flight route, the
airplane covered a specific geographical area during the investigation time. To account for the spacial variability we average the results of the fine resolution simulations over a sample area which encompasses the airplane route (Figure 4.1). In the following we particularly focus on investigating the performance of the model to represent the vertical structure of the clouds.

4.5.1 Case A: liquid-phase cloud on 19 April 2008 under polluted aerosol conditions

Case description
The weather regime on 19 April was characterized by a strong omega block and a ridge passing through central Alaska (McFarquhar et al., 2010). This omega block engendered deep convective systems associated with shortwave troughs propagating around the ridge. High aerosol concentrations observed on 19 April after the passage of a shortwave trough could be associated with biomass burning aerosols originating from agricultural and forest fires in northern Kazakhstan and southern Siberia (Warneke et al., 2009). The sampling period between 0:37 UTC and 02:50 UTC on 20 April was characterized by a low-level liquid cloud layer under polluted aerosol conditions.

Model results
For case A we conduct three different simulations with varying complexity in the treatment of aerosol-cloud interactions: a simulation with the standard aerosol module (STD), a simulation including all scavenging processes (SCAV) and a simulation accounting for aerosol processing in clouds including aerosol regeneration upon evaporation / sublimation (AP).

The three simulations feature nearly identical vertical profiles of temperature and relative humidity averaged over the entire sampling area (Fig.4.4). All observations obtained during the sampling period are vertically interpolated to the model levels and included in the compilation of the observed average vertical profiles. Both observations and model simulations display a comparable vertical temperature profile (Fig.4.4a) with decreasing temperatures in the lower 1000 m and rather constant temperatures higher up. However, at an altitude of about 1000 m, the sample domain averaged temperatures reveal a stronger and about 2 degrees colder temperature inversion than observed, whereas the observations show a smoother temperature change at this altitude. Above the inversion, the modeled profiles keep the cold bias of about 2 degrees, whereas below the inversion the temperature lapse rate is simulated within the observational uncertainty. The differences may be due to model deficiencies or inaccuracies in the driving ECMWF reanalysis data. Further, the interpolation of the observations may possibly
be biased as the measurements of different altitudes were taken at varying locations. Taking this into account, the observed temperature profile is reasonably well reproduced in all three simulations. The observed relative humidity profile is reproduced mostly within the standard deviation by the three simulations (Fig. 4.4 b). Though, the model simulates a sharp peak maximum value at an altitude of about 900 m which is higher and more pronounced than in the observations. The observed relative humidity is probably underestimated in the presence of the cloud between 400 and 1400 m.

Like in the observations, the model simulates warm-phase clouds in the concerned model area. In the simulations, we differentiate between cloudy and clear-sky model conditions based on a threshold liquid water content (LWC) of $10^{-5}$ g m$^{-3}$. For the observations, we apply the cloud phase data to identify in-cloud measurements. Figure 4.5 shows the in-cloud averaged vertical profiles of LWC. In the model, the maximum value of liquid water content is produced at an altitude between 900 and 1100 m, corresponding to the altitude of the temperature inversion and maximum relative humidity. The simulated maximum liquid water content of about 0.2 g m$^{-3}$ underestimates the peak value of the observations. However, the cloud top and cloud base are well represented in the model, leading to a pertinent simulation the vertical extent of the liquid cloud layer.

The simulation with aerosol processing allows to follow aerosol particles even when scavenged into hydrometeors. Figure 4.6 shows the averaged vertical profiles of the aerosol number transfer rates between the unactivated and in-hydrometeor aerosol modes in the model sample.
4.5. Comparison to observations

Figure 4.5: In-cloud averaged vertical profiles of the liquid water content (LWC) for case A (19 April 2008). In-cloud observations are vertically interpolated to the model levels with the standard deviation shaded in gray.

area. In the simulation, mainly activation scavenging along with in-cloud collision scaveng-

Figure 4.6: Vertical profiles of the aerosol number transfer rates between unactivated and in-hydrometeor aerosol modes averaged over the sample area between 0:30 UTC and 3:00 UTC for case A (19 April 2008). Only transfer rates larger than 100 particles m\(^{-3}\) s\(^{-1}\) are shown.

ing between cloud droplets and interstitial aerosol particles are responsible for the transfer of aerosol particles from the unactivated modes into the in-hydrometeor modes. Most of the re-

generated aerosol particles upon droplet evaporation are released in the accumulation mode and a smaller number also in the Aitken mode, while the number of newly formed coarse mode particles remains negligible. The total number of released aerosol particles is smaller than
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The number of scavenged particles, indicating that cloud microphysical processes and collision scavenging lead to a reduction of the cloud droplet number concentration and consequently to an accumulation of aerosol mass in the cloud droplets. However, the simulation does not present a steady state of the atmosphere, so that an enhanced formation of cloud droplets compared to cloud droplet evaporation increasing the total cloud amount is also conceivable.

The three simulations differ regarding the treatment of aerosol-cloud interactions. Therefore, the simulated clouds exert different effects on the aerosol population. This becomes apparent in the vertical profile of the aerosol number size distributions (Fig. 4.7) distinguishing between total aerosol under clear sky conditions and interstitial aerosol comprising only unactivated aerosol particles within clouds. To differentiate between clear-sky and cloudy conditions, we employ the provided cloud phase data to the observations. In the simulations, the threshold LWC of $10^{-5} \text{ g m}^{-3}$ is employed to distinguish between total clear-sky and interstitial aerosols.

The observations (Fig. 4.7a and e) show that the total aerosol population is dominated by the accumulation mode with only a negligible contribution of the coarse mode. Within the clouds, the interstitial aerosol particles reveal a small reduction of the accumulation mode in favor of

![Figure 4.7: Clear-sky and in-cloud averaged vertical profiles of aerosol number size distribution ($dN/d\ln r$) in cm$^{-3}$ distinguishing between total aerosol under clear sky conditions and interstitial aerosol comprising only unactivated aerosol particles within clouds for case A (19 April 2008) of PCASP measurements (observed, a and e), the standard simulation (STD, b and f), the simulation with scavenging processes (SCAV, c and g) and the simulation with aerosol processing (AP, d and h).](image-url)
the formation of coarse mode particles. The standard simulation STD (Fig. 4.7b and f) cannot reproduce these features, as the aerosol population is not affected by cloud processes leading to similar aerosol size distributions of the total and interstitial aerosol. A reduction of the number concentration of aerosol particles can be seen in simulation SCAV (Fig. 4.7c and g), where aerosol particles are removed by activation and collision scavenging. In simulation AP, a part of the scavenged aerosol particles is released back to the atmosphere by evaporation of droplets increasing the number concentration of unactivated aerosol particles compared to SCAV (Fig. 4.7d and h). As already shown in Figure 4.6, most of the generated aerosol particles are released back in the accumulation mode. Therefore, simulation AP does not reproduce the observed increase in coarse mode particles. In the simulations, the differences between the total clear-sky aerosol size distributions and the interstitial aerosol size distributions are less pronounced than in the observations. The differentiation between clear-sky and cloudy is based on the instantaneous atmospheric conditions. Therefore, clear-sky aerosol most likely also includes preliminarily processed air masses. Due to aerosol sedimentation and turbulent mixing, the vertical structure of the aerosol size distributions is blurred.

4.5.2 Case B: mixed-phase cloud on 26 April 2008 under clean aerosol conditions

Case description
On flight 31 on 26 April 2008 a single-layer mixed-phase stratocumulus cloud was observed in the vicinity of Barrow over the broken sea ice. A high pressure system was located over the Arctic Ocean inducing the formation of a stratocumulus deck north of Barrow (McFarquhar et al., 2010) that persisted for approximately 15 h. In-situ measurements were taken about 80 km north-northeast of Barrow, including several flight legs above, below and within the clouds layer. The sampling period of the observations between 0:35 UTC and 3:03 UTC on 27 April was characterized by low aerosol concentrations.

Model results
The sample domain averaged vertical temperature and relative humidity profiles are shown in Figure 4.8. For both profiles, the peak values of minimum temperature and maximum relative humidity are simulated at an altitude between 700 and 800 m (Fig. 4.8a), which corresponds to the altitude of the observed peak values. However, the simulations underestimate the value of minimum temperature possibly due to model deficiencies, inaccuracies in the driving boundary data or the applied interpolation method of the observational data.

The slope of the vertical relative humidity profile is not well represented in all three simula-
tions (Fig. 4.8b). Moreover the values of relative humidity are significantly underestimated in simulation STD. We suppose that precipitation formation during previous time steps leads to a considerable reductions of relative humidity in the model. The major differences between the simulations are a result of the different treatments of the aerosol scavenging and regeneration processes engendering variations in the number of interstitial aerosol particles and thus available CCN in the simulations. Accordingly, simulations STD may have experienced more precipitation formation than the other simulations.

**Figure 4.8:** Sample domain averaged vertical profiles of (a) temperature and (b) relative humidity for case B (26 April 2008). Observations are vertically interpolated to the model levels with the standard deviation shaded in gray.

**Figure 4.9:** In-cloud averaged vertical profiles of the (a) liquid water content and (b) ice water content for case B (26 April 2008). In-cloud observations are vertically interpolated to the model levels with the standard deviation shaded in gray.
Vertical profiles of in-cloud averaged liquid water content (LWC) and ice water content (IWC) are displayed in figure 4.9. We employed a threshold value of $10^{-5}$ g m$^{-3}$ to compile in-cloud averaged vertical profiles of LWC and IWC for the simulations. In-cloud measurements were identified based on the provided cloud phase data. The observations indicate the presence of a supercooled liquid cloud layer from which ice particles are sedimenting. The model revealed to be able to reproduce this characteristic vertical structure of Arctic mixed-phase clouds. The simulated values of liquid and ice water content are comparable to the observational data (Fig. 4.9a). Simulation AP is able to reproduce the observed cloud top, but slightly overestimates the peak LWC. Simulation STD better reproduces the observed LWC, but underestimates the cloud top height. However, simulation SCAV overestimates not only the vertical extent of the liquid cloud layer, but also liquid water content within the cloud leading to an enhancement of the liquid phase compared to the observations. Regarding the ice phase, the simulations reproduce the observed amount of cloud ice (Fig. 4.9b). However, there are differences regarding the vertical extent of the ice phase of the cloud. Simulation STD simulates ice down to an altitude of about 300 m. In simulation AP and SCAV, the ice phase can be discerned even below 300 m altitude. This is in better agreement to the observations, which identified ice crystals down to the lowest measurement points.

![Figure 4.10](image.png)

**Figure 4.10:** Vertical profiles of the aerosol number transfer rates between unactivated and in-hydrometeor aerosol modes averaged over the sample area between 0:30 UTC and 3:00 UTC for case B (26 April 2008). Only transfer rates larger than 10 particles m$^{-3}$ s$^{-1}$ are shown.

Figure 4.10 depicts the vertical profiles of the aerosol number transfer rates between unactivated and in-hydrometeor aerosol modes for the simulation with aerosol processing in the sample area. Similar to the liquid cloud in case A, most of the aerosol particles are transferred
to the in-hydrometeor modes via activation scavenging and a smaller part via in-cloud collision scavenging. In the mixed-phase simulation regenerated aerosol particles upon evaporation / sublimation are almost entirely released back in the accumulation mode.

Figure 4.11: Clear-sky and in-cloud averaged vertical profiles of aerosol number size distribution ($dN/dlnr$) in cm$^{-3}$ distinguishing between total aerosol under clear sky conditions and interstitial aerosol comprising only unactivated aerosol particles within clouds for case B (26 April 2008) of PCASP measurements (observed, a and e), the standard simulation (STD, b and f), the simulation with scavenging processes (SCAV, c and g) and the simulation with aerosol processing (AP, d and h).

The effect of the regenerated aerosol particles on the aerosol population becomes apparent in Figure 4.11 showing the vertical profile of the aerosol size distributions. During the analyzed time period, most observations were made within the cloud, leading to a limited availability of clear sky aerosol data. Regarding the interstitial aerosol, the observations reveal an important decline in accumulation mode particles at the altitude of the liquid cloud layer. At the same time, the concentrations of coarse mode particles are enhanced suggesting the growth of accumulation mode particles towards coarse mode sizes (Fig. 4.11a and e). However, the observed coarse mode particles may also originate from other sources. Similar to case A, the standard simulation (STD) is not able to reproduce any changes in the aerosol size distribution due to cloud microphysical processes (Fig. 4.11b and f). In simulation SCAV the impact of the scavenging processes becomes apparent as aerosol concentrations are significantly reduced within the liquid layer. This decline is observable in both, the total and the interstitial aerosol size distributions (Fig. 4.11c and g). In simulation AP, a considerable part of the scavenged
aerosol particles is replenished by aerosol regeneration upon evaporation/sublimation. As already seen in Figure 4.10, almost all newly released aerosol particles are within the size range of accumulation mode particles. Therefore, no significant increase in coarse mode particles can be detected in this simulation (Fig. 4.11d and h).

4.5.3 Sensitivity tests for the mixed-phase clouds on 26 April 2008

In the following section we modified the ice nuclei properties of the aerosol particles in the model in order to evaluate the impact of IN on the simulated cloud structure.

Different dust properties

Within the aerosol module dust can be considered either as internally mixed, soluble or as insoluble. The assumed mixing state impacts its potential to act as a cloud condensation or ice nuclei. We conducted simulations with the aerosol processing scheme in which we initially attributed the dust to the soluble modes (AP, identical to simulation AP in section 4.5.2), to the insoluble modes (AP DU-ins) and where we multiplied the insoluble dust amount by 100 (AP DU-ins100). Figure 4.12 displays the in-cloud averaged vertical profiles of the liquid water and ice water content for the different simulations. All simulation slightly overestimate the maximum liquid water content in the cloud layer (Fig. 4.12a). However, simulation AP and AP DU-ins100 adequately reproduce the cloud top altitude, but somewhat underestimate the altitude of the cloud base. In simulation AP DU-ins, the vertical LWC profile does not show such a sharp

Figure 4.12: In-cloud averaged vertical profiles of the (a) liquid water content and (b) ice water content for case B (26 April 2008). Dust is considered to be either internally mixed, soluble (AP) or insoluble (AP DU-ins) and is multiplied by 100 (AP DU-ins100). In-cloud observations are vertically interpolated to the model levels with the standard deviation shaded in gray.
maximum, but rather reveals the formation of a uniform liquid cloud layer with a significantly increased vertical extent as compared to the observations. Considering insoluble dust particles which can act as contact ice nuclei for the formation of ice crystals in AP DU-ins and AP DU-ins100 increases the vertical extent of the IWC better reproducing the observations (Fig. 4.12b). When multiplied by 100, insoluble dust particles (AP DU-ins) considerably affect the ice water content by enlarging the vertical extent of the ice phase as heterogeneous freezing processes become more important. Therefore, the AP simulation with enhanced dust loading ameliorates the representation of the observed mixed-phase cloud in the model.

**Bacteria as ice nuclei**

Bioaerosols, especially bacteria are found to be highly efficient ice nuclei. The current model version does not include bioaerosols. Assuming that all dust particles would have freezing efficiencies corresponding to those of bacteria, we estimate an upper limit of the effect of bioaerosols on the cloud properties. The freezing coefficients of dust are replaced by the freezing coefficients for bacteria given in Diehl and Wurzler (2004) and Diehl et al. (2006) (Table 4.5.3). We further distinguish between internally mixed, soluble bacteria (AP bacteria) and insoluble bacteria (AP bacteria-ins).

<table>
<thead>
<tr>
<th>Aerosol type</th>
<th>(a) [K(^{-1})]</th>
<th>(b) [m(^{-3})]</th>
<th>(c_1) [K(^{-1})]</th>
<th>(c_0)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bacteria</td>
<td>1.0</td>
<td>6.19 (\times 10^6)</td>
<td>26.41 (\times 10^{-2})</td>
<td>74.23 (\times 10^{-2})</td>
</tr>
</tbody>
</table>

**Table 4.2:** Material specific coefficients for bacteria, \(a\) and \(b\) for the parameterization of immersion freezing taken from table 2 in Diehl and Wurzler (2004), \(c_1\) and \(c_0\) for the parameterization of contact freezing taken from table 3 in Diehl et al. (2006).

The in-cloud averaged liquid and ice water content profiles of simulation AP (identical to simulation AP in section 4.5.2), AP bacteria and AP bacteria-ins are depicted in Fig. 4.13. Considering internally mixed, soluble bacteria (AP bacteria), the liquid water content is significantly decreased, whereas the simulation with insoluble bacteria (AP bacteria-ins) better represents the vertical profile and maximum values of LWC (Fig. 4.13a). However, both simulations underestimate the maximum LWC and altitude of the cloud as generated from the observations. Regarding the ice water content, both simulations with bacteria show a significant enhancement as compared to simulation AP (Fig. 4.13b). The consideration of bacteria as possible IN for ice formation considerably increases the cloud ice. Appropriate model performance regarding the vertical profiles of both LWC and IWC is obtained for insoluble bacteria which may act as contact IN in the simulations. When cycled through the cloud, the processed bacteria is considered internally mixed and may act as CCN or IN in the immersion freezing mode.
4.6 Summary and conclusions

In the present work we conducted two case studies of Arctic low-level liquid and mixed-phase clouds in order to evaluate the impact of aerosol processing in clouds on the simulated cloud properties. The focus of this work lies on the vertical structure of the clouds and aerosol population. We compared simulations of different complexity in the treatment of aerosol-cloud interactions with observational data from the ISDAC flight campaign which took place in Alaska in 2008.

In general, the observed vertical cloud structures were adequately simulated by the model simulations. In the case study of a liquid-phase cloud under polluted aerosol conditions the alteration of the aerosol population due to aerosol-cloud interactions has no significant effect on the simulated cloud properties. On the one hand, this may be due to a strong dynamical forcing of this specific case, or the system may be saturated with respect to aerosol particles and is therefore insensitive to the slightly modified aerosol population. Regarding the simulation of mixed-phase Arctic clouds under clean aerosol conditions, the results reveal a sensitivity to the treatment of aerosol-cloud interactions and to the assumed properties of potential ice nuclei. However, almost all simulations succeed in simulating a liquid cloud layer with precipitating ice crystals.

Figure 4.13: In-cloud averaged vertical profiles of the (a) liquid water content and (b) ice water content for case B (26 April 2008). AP represents the simulation with aerosol processing from section 4.5.2. Bacteria are considered to be either soluble, mixed (AP bacteria) or insoluble (AP bacteria-ins). In-cloud observations are vertically interpolated to the model levels with the standard deviation shaded in gray.
Both case studies clearly show that aerosol processing in clouds releases a substantial amount of the scavenged aerosol particles back to the atmosphere. The observations clearly indicate augmented coarse mode particles in the interstitial aerosol. However, the aerosol processing scheme mainly generates aerosol particles in the accumulation mode, and only scarcely in the coarse mode. This may be due to a simplified representation of the involved aerosol microphysical and cloud microphysical processes or a too short residence time within the hydrometeors. For the employed aerosol processing scheme, the involved cloud microphysical processes are crucial. In order to form coarse mode particles, a considerable accumulation of aerosol mass in the droplets is essential, often involving multiple collision processes. It is also important to mention that the reality of the high number concentration of coarse mode particles remains questionable, as within the cloud, the PCASP measurements can be influenced by cloud hydrometeors, artificially increasing the number of observed particles.

The sensitivity test show, that enhanced contact freezing allows to better reproduce the observed vertical profile of the IWC. Contact freezing may be increased due to additional contact IN as simulated for insoluble dust. Considering insoluble bacteria as contact IN in the simulation increases also the freezing efficiency in the contact mode allowing for a better representation of the LWC and IWC. These results illustrate, that the relation between IN number concentration and properties, the involved freezing modes and the ice phase of the cloud are still not well understood.

Our results show, that the consideration of scavenging processes without accounting for aerosol regeneration (simulation SCAV) does not ameliorate the representation of the clouds and rather degrades the representation of the aerosol size distribution. Furthermore, there are only minor differences between the simulation with aerosol processing and the standard simulation suggesting that the present case studies are not IN-limited. Based on these results, a clear superiority of one of the model versions cannot be detected. However, the new aerosol processing scheme still adequately simulated the observed liquid and mixed-phase cloud demonstrating that the aerosol processing scheme represents an appropriate tool to study aerosol-cloud interactions.
Chapter 5

Summary and outlook

5.1 Summary

In order to improve our understanding of aerosol-cloud interactions and their effects on aerosol and cloud properties, a new modeling framework based on the bulk aerosol microphysics module M7 (Vignati et al., 2004) and the two-moment cloud scheme of Seifert and Beheng (2006) was introduced into the regional weather forecast and climate model COSMO (Doms and Schättler, 2002; Steppeler et al., 2003). The new modeling framework allows to simulate not only aerosol effects on clouds, but also cloud effects on aerosol particles, summarized within the term "aerosol processing in clouds". For this, five new aerosol modes for in-hydrometeor aerosol particles were introduced into the model, corresponding to the five prognostic cloud hydrometeor classes of the cloud microphysical scheme which are cloud droplets, ice crystals, rain drops, snow flakes and graupel. The distinction between unactivated and in-hydrometeor aerosol particles enables the simulation of cloud cycling of aerosol particles. Due to cloud droplet activation, heterogeneous ice nucleation and collision-coalescence processes aerosol particles are incorporated into hydrometeors where they are subject to cloud microphysical processes. They are released back to the atmosphere upon evaporation and sublimation of hydrometeors. The newly developed extension of the aerosol module M7 is based on the approach by Hoose et al. (2008a) developed for the treatment of aerosol processing in the global climate model ECHAM-HAM. A first part of the work was dedicated to the development and implementation of the new extension of the modeling system. The model physics were presented in Chapter 2.
Chapter 5. Summary and outlook

5.1.1 Microphysical processing of aerosol particles in orographic clouds

In an idealized 2D study we have investigated the effects of liquid and mixed-phase orographic clouds on the aerosol population. The simulations showed that the aerosol population is greatly affected by aerosol scavenging processes as well as by aerosol processing in clouds including the regeneration of newly formed aerosol particles upon evaporation and sublimation. The processes not only affect the total aerosol number concentrations, but also alter the shape of the aerosol size distributions. Accounting for aerosol regeneration upon evaporation / sublimation partly replenishes the scavenged aerosol particles. At this, the formation of coarse mode particles is observed near to the surface, whereas at higher altitudes mainly accumulation mode particles are produced. Due to the transport within sedimenting hydrometeors, a vertical redistribution of aerosol number and mass occurs for both the liquid and mixed-phase clouds. Precipitating snow flakes efficiently remove aerosol mass from the atmosphere.

The study also allowed to identify different aerosol cycles for the clouds. In the simulated non-precipitating liquid cloud aerosol mass can be found in cloud droplets and rain drops. At this, activation scavenging, autoconversion and evaporation of cloud droplets and rain drops are primarily responsible for the transfer of aerosol mass. In the simulated precipitating mixed-phase cloud, two aerosol cycles were identified. A first cycle includes activation scavenging and cloud droplet evaporation due to the WBF process. Being connected to the first cycle via the riming process, a second cycle comprises the interactions with snow flakes. Significant amounts of aerosol mass are primarily found in cloud droplets and snow flakes. Ice crystals are formed heterogeneously from a few cloud droplets allowing only for a limited transfer of aerosol mass.

Regenerated internally mixed, soluble accumulation and coarse mode particles add to the number of potential CCN in the model which in turn increases the cloud droplet number concentration upon subsequent cloud formation. In the simulated mixed-phase cloud, an influence on the ice crystal number concentration is also observable via the changes in cloud droplet number concentration. However, the impact on the ice water content remains negligible as most of the cloud ice is formed via the WBF process which appears to be independent from the number of preexisting ice particles in the present setup.

5.1.2 Simulating Arctic springtime low-level clouds

In two case studies the model's ability to simulate the vertical structure of Arctic low-level liquid and mixed-phase clouds was demonstrated. Due to aerosol processing a significant part of
the scavenged aerosol particles is regenerated. However, the new aerosol particles are mostly released to the accumulation mode, omitting the formation of numerous coarse mode particles as seen in the observations. Sensitivity studies regarding the IN properties show that enhanced contact freezing allows to ameliorate the representation of the vertical profile of LWC and IWC in the simulation with aerosol processing. At this, additional dust contact IN or the consideration of bacteria as contact IN may be important.

5.1.3 Outlook

In order to advance the research of aerosol-cloud interactions, different extensions of this work are recommendable.

**Observational data**

The presented 2D study of aerosol processing in orographic clouds does not include a comparison of the model results with atmospheric observations. Measurements of the aerosol properties upslope and downslope of a mountain with an orographic cloud, as well as of the cloud properties over the mountain are adequate to evaluate the model performance in such a setup. Accordant measurements were recently collected during the Hill Cap Cloud Thuringia 2010 campaign in Germany (e.g. Henning et al., 2014). Furthermore, detailed measurements of the aerosol composition and size distribution of the total and interstitial, and also the in-cloud aerosol particles during other field campaigns would give valuable information about the processing of aerosol particles and allow to better assess the performance of the models.

**Cloud microphysical parameterizations**

The present aerosol processing scheme is closely coupled to the cloud microphysical scheme of Seifert and Beheng (2006). Any improvement of the parameterizations used within this scheme therefore also has an impact on the representation of aerosol processing in the clouds. An example would be the implementation of a more accurate parameterization for the activation of cloud droplets or the advancement of the freezing parameterizations.

**Giant cloud condensation nuclei**

The newly generated aerosol particles released upon evaporation / sublimation may have an increased size compared to the original one due to prior aerosol processing within the cloud. Some of the very large particles are possible candidates for giant cloud condensation nuclei (GCCN) which encourage rapid rain formation in warm-phase clouds (Saleeby et al., 2009; Posselt and Lohmann, 2008). This may have implications for the precipitation rate and lifetime.
of the cloud. The implementation of a specific treatment of GCCN is a further extension of this work.

**Bioaerosols as IN**

Laboratory studies suggest, that bioaerosols like pollen, leaf litter, bacteria and spores can be very efficient ice nuclei especially in the contact- and immersion mode (e.g. Szyrmer and Zawadzki, 1997; Diehl et al., 2006). However, their effect on clouds and precipitation is still unknown on different scales, as the current knowledge of their emissions and lifetimes is very limited. Global simulations suggest that bacteria have only a minor influence on the Earth’s climate (Hoose et al., 2010; Sesartic et al., 2012), but they still might have important regional effects, especially in highly vegetated regions. A further possibility of extending this work is therefore the implementation of bioaerosols, especially bacteria, as potential IN in the model. A possible deactivation of these aerosols in the contact freezing mode due to cloud processing might be investigated with the aerosol processing scheme.

**Coupling to chemistry**

The present aerosol processing scheme focuses on cloud microphysical processing of aerosol particles in clouds. However, chemical reactions within cloud droplets or absorption of gases into the droplets may also significantly modify the amount and composition of the incorporated aerosol mass with implications for the properties of the released aerosol particles. The coupling of the aerosol processing module with a chemical model in order to consider both physical and chemical processing of aerosol particles is a further extension of this work.

**High resolution studies**

Another extension represents the setup of large eddy simulation studies, allowing for a detailed investigation of each microphysical process and its impact on aerosol processing in clouds. Paukert and Hoose (2014) successfully conducted high resolution simulations with the regional model COSMO focusing on the evaluation of a new immersion freezing parameterization by a case study of Arctic mixed-phase clouds. Regarding aerosol processing in clouds a detailed process study may give valuable insights in the ponderousness of the different processes.
# List of Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D</td>
<td>two-dimensional</td>
</tr>
<tr>
<td>3D</td>
<td>three-dimensional</td>
</tr>
<tr>
<td>AERONET</td>
<td>Aerosol Robotic Network</td>
</tr>
<tr>
<td>AP</td>
<td>Aerosol particle</td>
</tr>
<tr>
<td>BC</td>
<td>Black carbon</td>
</tr>
<tr>
<td>CCN</td>
<td>Cloud condensation nuclei</td>
</tr>
<tr>
<td>CD</td>
<td>In-cloud droplet aerosol mass mode</td>
</tr>
<tr>
<td>COSMO</td>
<td>Consortium of Small-scale MOdeling</td>
</tr>
<tr>
<td>COSMO model</td>
<td>Regional weather forecast and climate model of the COSMO consortium</td>
</tr>
<tr>
<td>CCLM</td>
<td>Climate version of the COSMO model</td>
</tr>
<tr>
<td>DU</td>
<td>Dust</td>
</tr>
<tr>
<td>DWD</td>
<td>German Weather Service (Deutscher Wetter Dienst)</td>
</tr>
<tr>
<td>ECHAM5-HAM</td>
<td>General circulation model derived from the ECMWF model, developed at the Max-Planck Institute in Hamburg, Germany</td>
</tr>
<tr>
<td>ECMWF</td>
<td>European center for medium range weather forecasts</td>
</tr>
<tr>
<td>GCCN</td>
<td>Giant cloud condensation nuclei</td>
</tr>
<tr>
<td>GR</td>
<td>In-graupel aerosol mass mode</td>
</tr>
<tr>
<td>IC</td>
<td>In-ice crystal aerosol mass mode</td>
</tr>
<tr>
<td>IN</td>
<td>Ice nuclei</td>
</tr>
<tr>
<td>IPCC</td>
<td>Intergovernmental Panel on Climate Change</td>
</tr>
<tr>
<td>ISDAC</td>
<td>Indirect and Semi-Direct Aerosol Campaign</td>
</tr>
<tr>
<td>IWC</td>
<td>Ice water content</td>
</tr>
<tr>
<td>LWC</td>
<td>Liquid water content</td>
</tr>
<tr>
<td>M7</td>
<td>Aerosol microphysics module</td>
</tr>
<tr>
<td>OC</td>
<td>Organic carbon</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Description</td>
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<tr>
<td>--------------</td>
<td>-------------</td>
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<tr>
<td>PCASP</td>
<td>Passive Cavity Aerosol Spectrometer Probe</td>
</tr>
<tr>
<td>POM</td>
<td>Particulate organic matter</td>
</tr>
<tr>
<td>QC</td>
<td>Cloud water mixing ratio</td>
</tr>
<tr>
<td>QI</td>
<td>Ice water mixing ratio</td>
</tr>
<tr>
<td>QNC</td>
<td>Number concentration of cloud droplets</td>
</tr>
<tr>
<td>QNI</td>
<td>Number concentration of ice crystals</td>
</tr>
<tr>
<td>QR</td>
<td>Rain water mixing ratio</td>
</tr>
<tr>
<td>QS</td>
<td>Snow water mixing ratio</td>
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<tr>
<td>RD</td>
<td>In-rain drop aerosol mass mode</td>
</tr>
<tr>
<td>SF</td>
<td>In-snow flake aerosol mass mode</td>
</tr>
<tr>
<td>SHEBA</td>
<td>Surface Heat Budget of the Arctic Ocean campaign</td>
</tr>
<tr>
<td>SMPS</td>
<td>Scanning mobility particle sizer</td>
</tr>
<tr>
<td>SS</td>
<td>Sea salt</td>
</tr>
<tr>
<td>SU</td>
<td>Sulfate</td>
</tr>
<tr>
<td>TKE</td>
<td>Turbulent kinetic energy</td>
</tr>
<tr>
<td>UTC</td>
<td>Coordinated universal time</td>
</tr>
<tr>
<td>WBF process</td>
<td>Wegener-Bergeron-Findeisen process</td>
</tr>
</tbody>
</table>
### List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$ (in eq.2.2)</td>
<td>Constant in hydrometeor diameter-mass relation</td>
<td>$m , kg^{-b}$</td>
</tr>
<tr>
<td>$a$ (in eq.2.11)</td>
<td>Parameter in freezing rate equation</td>
<td>$K^{-1}$</td>
</tr>
<tr>
<td>$a$ (in eq.3.7)</td>
<td>Mountain half-width</td>
<td>$m$</td>
</tr>
<tr>
<td>$a$ (in eq.3.8)</td>
<td>Parameter of the idealized relative humidity profile</td>
<td></td>
</tr>
<tr>
<td>$A$</td>
<td>Coefficient in the generalized $\Gamma$-distribution</td>
<td>$kg^{-\nu-1} , m^{-3}$</td>
</tr>
<tr>
<td>$b$ (in eq.2.2)</td>
<td>Constant in hydrometeor diameter-mass relation</td>
<td></td>
</tr>
<tr>
<td>$b$ (in eq.2.11)</td>
<td>Ice nucleation efficiency in freezing rate equation</td>
<td>$m^{-3}$</td>
</tr>
<tr>
<td>$b$ (in eq.3.8)</td>
<td>Parameter of the idealized relative humidity profile</td>
<td>$m^{-1}$</td>
</tr>
<tr>
<td>$B_{\text{subst}}$</td>
<td>Material-specific, but size-dependent freezing efficiency of the aerosol component $\text{subst}$</td>
<td>$m^{-3}$</td>
</tr>
<tr>
<td>$c$ (in eq.3.8)</td>
<td>Parameter of the idealized relative humidity profile</td>
<td>$m^{-1}$</td>
</tr>
<tr>
<td>$c_{\text{turb}}$</td>
<td>Parameter for the updraft vertical velocity</td>
<td></td>
</tr>
<tr>
<td>$C$</td>
<td>Cunningham slip correction factor</td>
<td></td>
</tr>
<tr>
<td>$D_a$</td>
<td>Aerosol diameter</td>
<td>$m$</td>
</tr>
<tr>
<td>$D_h$</td>
<td>Diameter of hydrometeors</td>
<td>$m$</td>
</tr>
<tr>
<td>$D_k$</td>
<td>Aerosol Brownian diffusivity</td>
<td>$m^2 , s^{-1}$</td>
</tr>
<tr>
<td>$E(D_a, D_h)$</td>
<td>Collection efficiency</td>
<td></td>
</tr>
<tr>
<td>$f_{\text{subst}}^*$</td>
<td>Normalized surface fraction of the aerosol component $\text{subst}$ which acts as an IN</td>
<td></td>
</tr>
<tr>
<td>$f_{\text{precip}}$</td>
<td>Fraction of precipitating rain or snow</td>
<td></td>
</tr>
<tr>
<td>$h_0$</td>
<td>Mountain peak height</td>
<td>$m$</td>
</tr>
<tr>
<td>$J_{\text{IFR}}$</td>
<td>Heterogeneous freezing rate</td>
<td>$m^{-3} , s^{-1}$</td>
</tr>
<tr>
<td>$K_{\text{hydro,k}}$</td>
<td>Collision kernel for hydrometeors of class $\text{hydro}$ with aerosols of mode $k$</td>
<td>$m^3 , s^{-1}$</td>
</tr>
<tr>
<td>$L_c$</td>
<td>Liquid water content (cloud droplets)</td>
<td>$kg , m^{-3}$</td>
</tr>
<tr>
<td>Symbol</td>
<td>Description</td>
<td>Unit</td>
</tr>
<tr>
<td>----------</td>
<td>-----------------------------------------------------------------------------</td>
<td>-----------------</td>
</tr>
<tr>
<td>$m$</td>
<td>Mass of hydrometeors</td>
<td>kg</td>
</tr>
<tr>
<td>$M_{k}^{\text{subst}}$</td>
<td>Aerosol mass concentration of compound $\text{subst}$ of the k-th aerosol mode</td>
<td>kg kg$^{-1}$</td>
</tr>
<tr>
<td>$N_{&gt;35\text{nm}}$</td>
<td>Number concentration of internally mixed, soluble aerosol particles larger than 35 nm</td>
<td>m$^{-3}$</td>
</tr>
<tr>
<td>$N_{\text{hydro}}$</td>
<td>Hydrometeor number concentration of class $\text{hydro}$</td>
<td>m$^{-3}$</td>
</tr>
<tr>
<td>$N_k$</td>
<td>Aerosol number concentration of the k-th aerosol mode</td>
<td>m$^{-3}$</td>
</tr>
<tr>
<td>$N_{k,\text{con}}$</td>
<td>Number concentration of potential contact IN in the aerosol mode k</td>
<td>m$^{-3}$</td>
</tr>
<tr>
<td>$p$</td>
<td>Pressure</td>
<td>Pa</td>
</tr>
<tr>
<td>$Q_{\text{hydro},k}$</td>
<td>Collision rate between hydrometeor class $\text{hydro}$ and aerosol mode k</td>
<td>m$^{-3}$ s$^{-1}$</td>
</tr>
<tr>
<td>$r$</td>
<td>Aerosol radius</td>
<td>m</td>
</tr>
<tr>
<td>$\bar{r}_k$</td>
<td>Count median radius of the k-th mode</td>
<td>m</td>
</tr>
<tr>
<td>$RH$</td>
<td>Relative humidity</td>
<td></td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
<td>s</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
<td>K</td>
</tr>
<tr>
<td>$U_t$</td>
<td>Terminal fall velocity</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>$v$</td>
<td>Terminal fall velocity of hydrometeors</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>$V_{\text{tot}}$</td>
<td>Volume of aerosol mass</td>
<td>m$^3$</td>
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<tr>
<td>$w$</td>
<td>Updraft vertical velocity</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>$w_g$</td>
<td>Grid-scale vertical velocity</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>$z$</td>
<td>Vertical coordinate (geometric height)</td>
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</tr>
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<td>$z_0$</td>
<td>Parameter of the idealized relative humidity profile</td>
<td>m</td>
</tr>
<tr>
<td>$\alpha$ (in eq.2.3)</td>
<td>Constant in relation for hydrometeor terminal fall velocities</td>
<td>m s$^{-1}$ kg$^{-\beta}$</td>
</tr>
<tr>
<td>$\alpha$ (in eq. 2.7)</td>
<td>Parameter in activation parameterization</td>
<td>m$^{-4}$ s$^{-1}$</td>
</tr>
<tr>
<td>$\beta$ (in eq.2.3)</td>
<td>Constant in relation for hydrometeor terminal fall velocities</td>
<td></td>
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<tr>
<td>Symbol</td>
<td>Description</td>
<td>Unit</td>
</tr>
<tr>
<td>--------</td>
<td>-----------------------------------------------------------------------------</td>
<td>---------------</td>
</tr>
<tr>
<td>( \gamma ) (in eq.2.3)</td>
<td>Constant in relation for hydrometeor terminal fall velocities</td>
<td></td>
</tr>
<tr>
<td>( \epsilon )</td>
<td>Freezing efficiency</td>
<td></td>
</tr>
<tr>
<td>( \lambda ) (in eq. 2.1)</td>
<td>Slope parameter in hydrometeor size distribution</td>
<td>kg(^{-\mu})</td>
</tr>
<tr>
<td>( \lambda )</td>
<td>Mean free path length</td>
<td>m</td>
</tr>
<tr>
<td>( \Lambda )</td>
<td>Scavenging coefficient</td>
<td></td>
</tr>
<tr>
<td>( \mu )</td>
<td>Constant in generalized G-distribution</td>
<td></td>
</tr>
<tr>
<td>( \nu )</td>
<td>Constant in generalized G-distribution</td>
<td></td>
</tr>
<tr>
<td>( \rho )</td>
<td>Density of air</td>
<td>kg m(^{-3})</td>
</tr>
<tr>
<td>( \rho_{\text{subst}} )</td>
<td>Density of aerosol component ( \text{subst} )</td>
<td>kg m(^{-3})</td>
</tr>
<tr>
<td>( \rho_{w} )</td>
<td>Density of water</td>
<td>kg m(^{-3})</td>
</tr>
<tr>
<td>( \sigma_k )</td>
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