Aerosol processing and its effect on mixed-phase clouds in a global climate model

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Aerosol Processing
and its Effect on Mixed-Phase Clouds
in a Global Climate Model

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for the degree of
Doctor of Sciences

presented by
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Abstract

Aerosol particles undergo substantial modifications in the atmosphere, between their emission or formation and their removal. Coagulation, coating, water uptake and surface chemistry alter the aerosol size distribution and composition. Clouds contribute significantly to the changes in the aerosol properties by physical and chemical processing inside cloud droplets. Cloud droplets nucleate on aerosol particles of suitable sizes and hygroscopicity, and subsequently collide with other droplets or interstitial particles. Chemical reactions in the aqueous phase can add non- or semi-volatile components to the cloudborne aerosol mass. Evaporating droplets release the collected material as larger, internally mixed particles. Droplet evaporation is of special importance in mixed-phase clouds, where the difference in saturation water vapor pressure between liquid and ice water leads to the growth of ice crystals on the expense of evaporating droplets (Wegener-Bergeron-Findeisen process). Furthermore, mixed-phase clouds in the temperature range between 0 and −35°C are sensitive to the number of aerosol particles acting as ice nuclei, which are required for droplet freezing. Some uncoated particles can act as contact ice nuclei at relatively high temperatures, while particles coated with a soluble material (e.g. by cloud processing) initiate ice formation rather in the immersion freezing mode at lower temperatures.

For simulation of aerosol processing in clouds, the global aerosol-climate model ECHAM5-HAM has been extended by an explicit treatment of cloud-borne particles. Two additional modes for in-droplet and in-crystal particles are introduced, which are coupled to the number of cloud droplet and ice crystal concentrations simulated by the ECHAM5 double-moment cloud microphysics scheme. Transfer, production and removal of cloud-borne aerosol number and mass by cloud droplet activation, collision scavenging, aqueous-phase sulfate production, freezing, melting, evaporation, sublimation and precipitation formation are taken into account. The model performance in single-column mode is demonstrated and validated with observations of the evolution of total and interstitial aerosol concentrations and size distributions during three different mixed-phase cloud events at the alpine high-altitude research station Jungfraujoch (Switzerland). Although the simulations can not be compared one-to-one with the observations, the governing processes in the evolution of the cloud and aerosol parameters are captured qualitatively well. High scavenged fractions are found during the presence of liquid water, while the release of particles during the Wegener-Bergeron-Findeisen process results in low scavenged fractions after cloud glaciation. The observed coexistence of liquid and ice, which might be related to cloud heterogeneity at subgrid scales, can only be simulated in the model when assuming non-equilibrium conditions.

The explicit treatment of in-cloud aerosol particles in ECHAM5-HAM allows a global assessment of the turnover of aerosol particles in stratiform clouds. Compared to previous estimates, ECHAM5-HAM simulates a lower number of cycles through clouds (on global average only 0.4), mainly because aerosol processing is, for the time being, limited to stratiform clouds. The rates of aerosol mass transfer into cloud droplets and ice crystals are analyzed, and it is found that nucleation scavenging dominates over collision scavenging for sulfate, sea salt and mineral dust, while the two processes are of similar importance for black carbon and organic carbon. Aqueous-phase formation of sulfate is the most important source of atmospheric sulfate particles. As in the simulation with explicit aerosol processing fewer particles are scavenged into the cloud phase than in the control simulation, more particles are available for activation, resulting in higher cloud droplet concentrations. Comparison to different observations reveals several inconsistencies. While the marine boundary layer accumulation mode number concentrations are better simulated with the new treatment, the opposite is true for the droplet concentrations in marine boundary layer clouds. Sea salt mass concentrations are well simulated over ocean, but overestimated over land. The wet and dry diameter of the accumulation mode is overestimated especially over ocean. Total volume burden and aerosol optical depth at a large number of stations is not well simulated. The scavenged particle number fraction at the Jungfraujoch is satisfactorily simulated in both simulations. While for the standard model this is analyzed only diagnostically and the scavenged fraction for wet deposition is fixed to excessively high values at low temperatures, the scavenged fraction in the new scheme is consistently applied for wet deposition calculations.

Arctic mixed-phase clouds are the focus of further single-column model studies, which reveal that heterogeneous ice nucleation is of crucial importance for the evolution of these clouds. ECHAM5-HAM simulates persistent supercooled liquid water clouds at temperatures below 0°C in accordance with ob-
servations, but insufficient ice crystal growth after freezing of a small fraction of the droplets. The results are very sensitive to the prescribed aerosol properties, for which only limited observations are available.

Finally, heterogeneous freezing in the contact and immersion mode on different aerosols is studied in the global model. Mineral dust is the dominant natural ice nucleating aerosol. Its ice nucleation efficiency depends on the mineralogical composition. We show the first sensitivity studies with a global climate model and a three-dimensional dust mineralogy. Results show that depending on the dust mineralogical composition, coating with soluble material from anthropogenic sources can lead to quasi-deactivation of natural dust ice nuclei. This effect counteracts the increased cloud glaciation by anthropogenic black carbon particles. The resulting aerosol indirect effect through the glaciation of mixed-phase clouds by black carbon particles is small.


Zusammenfassung

ren am Jungfraujoch wird in beiden Simulationen gut wiedergegeben. Während diese Analyse für das Standardmodell jedoch rein diagnostisch ist und für die nasse Deposition bei niedrigen Temperaturen zu viele Aerosole ausgewaschen werden, wird der ausgewaschene Anteil in dem neuen Prozessierungsschema konsistent auch für die nasse Deposition angewendet.

Arktische Mischwolken stehen im Zentrum von weiteren Säulenmodellstudien, die zeigen, dass heterogene Eisnukleation für die Entwicklung dieser Wolken von zentraler Bedeutung ist. ECHAM5-HAM simuliert langlebiges unterkühltes Wolkenwasser bei Temperaturen unter 0°C, was auch von den Beobachtungen bestätigt wird. Das Wachstum der Eiskristalle nach dem Gefrieren einiger weniger Tropfen wird jedoch unterschätzt. Die Ergebnisse hängen sehr stark von den vorgeschriebenen Aerosoleigenschaften ab, für die es nur wenige Messungen gibt.

Contents

Abstract

Zusammenfassung

1 Introduction: Aerosol effects on clouds and cloud effects on aerosols
  1.1 The atmospheric aerosol .................................................. 1
  1.2 Aerosol indirect effects on climate via clouds .......................... 2
    1.2.1 Aerosol indirect effects via liquid clouds ......................... 3
    1.2.2 Aerosol indirect effects via mixed-phase clouds .................. 4
  1.3 Aerosol-cloud interactions in global climate models .................. 5
    1.3.1 Global aerosol modeling ............................................ 5
    1.3.2 Aerosol-cloud coupling ............................................ 6
    1.3.3 ECHAM5-HAM ......................................................... 8
    1.3.4 Mixed-phase clouds in ECHAM5-HAM ................................. 9
  1.4 Overview over this dissertation ....................................... 10

2 Aerosol processing in mixed-phase clouds in ECHAM5-HAM: Model description and comparison to observations
  2.1 Introduction .............................................................. 11
  2.2 Model description ........................................................ 13
    2.2.1 ECHAM5 .............................................................. 13
    2.2.2 Introducing cloud-borne modes in ECHAM5-HAM ..................... 13
    2.2.3 SCM model setup .................................................... 18
  2.3 Observations of in-cloud and interstitial aerosol size distributions during CLACE 3 19
  2.4 SCM results for CLACE mixed-phase clouds ................................ 20
  2.4.1 Case A: liquid cloud (March 30) .................................... 20
  2.4.2 Case B: glaciating cloud (March 10) .................................. 24
  2.4.3 Case C: coexistence of liquid and ice (March 20) ................... 28
  2.5 Summary and conclusions ............................................... 30

3 Global simulations of aerosol processing in clouds
  3.1 Introduction .............................................................. 33
  3.2 Model description ........................................................ 33
  3.3 Comparison to the standard model ..................................... 35
    3.3.1 Global and zonal mean cloud and aerosol parameters ............. 35
  3.4 In-cloud aerosol budgets ............................................... 37
    3.4.1 Scavenged aerosol mass .............................................. 37
    3.4.2 Transfer rates between in-cloud and interstitial modes .......... 40
  3.4.3 Life cycles of cloud condensate and in-cloud aerosol ............ 40
  3.5 Comparison with observations .......................................... 44
    3.5.1 Marine boundary layer aerosol .................................... 44
    3.5.2 Sea salt mass concentrations .................................... 44
    3.5.3 AERONET size distributions .................................... 46
    3.5.4 AERONET aerosol optical depth .................................. 50
    3.5.5 Marine boundary layer cloud droplets ............................. 50
    3.5.6 Scavenged fraction at Jungfraujoch ............................... 53
    3.5.7 Sensitivity studies on cloud droplet activation .................. 57
  3.6 Conclusions ............................................................. 60
### Contents

4 **Modelling of Arctic mixed-phase clouds: M-PACE**  
  4.1 Arctic clouds ............................................. 63  
  4.2 The Mixed-Phase Arctic Cloud Experiment (M-PACE) ........ 64  
  4.3 Period A .................................................... 65  
  4.4 Period B .................................................... 66  

5 **Global influence of dust mineralogical composition on heterogeneous ice nucleation in mixed-phase clouds**  
  5.1 Introduction ............................................... 71  
  5.2 Model Description ......................................... 72  
    5.2.1 Heterogeneous Freezing Parameterization ................ 72  
    5.2.2 Dust Mineralogical Composition ....................... 75  
    5.2.3 Evaluation of the Dust Mineralogical Composition Fields ........................................... 77  
    5.2.4 Simulations .......................................... 77  
  5.3 Results .................................................... 79  
    5.3.1 Simulations of present-day climate ................... 79  
    5.3.2 Anthropogenic Climate Forcing ......................... 81  
    5.3.3 Sensitivity to freezing on black carbon ............... 84  
  5.4 Discussion and conclusions ................................ 84  

6 **Summary and outlook** ........................................ 89  
  6.1 Summary ................................................... 89  
  6.2 Future work ................................................ 89  
    6.2.1 Collision scavenging .................................. 89  
    6.2.2 Aerosol processing in convective clouds ............... 90  
    6.2.3 Coupling freezing parameterizations to the aerosol processing scheme ...................... 90  
    6.2.4 Biogenic ice nuclei .................................... 92  

List of symbols and acronyms ....................................... 93  

References .................................................................. 97  

Curriculum vitae .................................................... 109  

Acknowledgements ................................................... 111
1 Introduction: Aerosol effects on clouds and cloud effects on aerosols

1.1 The atmospheric aerosol

Small solid and liquid airborne particles, visible to the eye only if their concentration is exceptionally high, have important effects on the earth’s radiation budget and on clouds. Strictly speaking, the term “aerosol” refers to a colloidal system in which the dispersed phase is composed of either solid or liquid particles, and in which the dispersion medium is some gas, usually air (Glickmann, 2000); but often the airborne particles alone are loosely referred to as “aerosols”. Aerosol particles can be emitted from natural sources, such as windblown desert dust, or they can be generated by human pollution. Often a distinction is drawn between primary and secondary aerosol particle formation. Primary aerosols are directly emitted as particles, e.g. desert dust, sea salt, pollen or industrial particle emissions. Secondary aerosols form in the atmosphere from gas-to-particle conversion, e.g. nucleation of sulfate particles at low temperatures in the upper troposphere. The size of aerosol particles can range from a few nanometers to tens of micrometers. Often different size ranges are referred to as “modes”, which are influenced by different regimes of production and removal. The smallest particles constitute the “nucleation mode”, formed from low-volatility gases. For these particles Brownian diffusion is efficient, and they rapidly grow by coagulation and condensation of more low-volatility material into the so-called “Aitken mode” (diameter \( \approx 10–100 \text{ nm} \)). Combustion particles fall also in the Aitken mode range. After further growth by coagulation and condensation, diffusion becomes less and less efficient, and the range between approximately 100 nm to 1 \( \mu \text{m} \) is called “accumulation mode”. Primary particles are often of larger sizes and constitute the “coarse mode”. The dominating removal process for the coarse mode is sedimentation. For accumulation mode particles neither sedimentation nor diffusion is efficient, and the dominant sink is in-cloud scavenging by cloud and particles (Seinfeld and Pandis, 1998). Table 1.1 summarizes the aerosol size modes.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Size range</th>
<th>Typical components</th>
<th>Dominant sink</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nucleation mode</td>
<td>( d \leq 10 \text{ nm} )</td>
<td>sulfate</td>
<td>growth by coagulation and condensation</td>
</tr>
<tr>
<td>Aitken mode</td>
<td>( 10 \text{ nm} &lt; d \leq 100 \text{ nm} )</td>
<td>sulfate, soot</td>
<td>growth by coagulation and condensation</td>
</tr>
<tr>
<td>Accumulation mode</td>
<td>( 100 \text{ nm} &lt; d \leq 1 \mu \text{m} )</td>
<td>all</td>
<td>in-cloud scavenging</td>
</tr>
<tr>
<td>Coarse mode</td>
<td>( 1 \mu \text{m} &lt; d \leq 10 \mu \text{m} )</td>
<td>sea salt, dust</td>
<td>sedimentation</td>
</tr>
<tr>
<td>Supercoarse mode</td>
<td>( 10 \mu \text{m} &lt; d )</td>
<td>sea salt, dust</td>
<td>sedimentation</td>
</tr>
</tbody>
</table>

The most important aerosol chemical components are:

- sea salt, emitted from the oceans through evaporation of sea spray
- mineral dust, wind-driven emission from deserts and disturbed soils
- sulfate, formed from oxidized precursor gases, which are emitted from the oceans, volcanoes, the biosphere, and anthropogenic pollution
- black carbon (soot), from biomass burning and anthropogenic pollution
- organic carbon, formed from biogenic or anthropogenic volatile organic compounds
- nitrate and ammonium (semi-volatile species), emitted from soils (precursor gases), fossil-fuel combustion, agriculture, biomass burning
- primary organic particles (pollen, bacteria, fungi ...)
• particulate emissions from industry and transport, like fly-ash, metallic aerosols, road abrasion particles

• meteoritic dust

A schematic view of aerosol processes in the atmosphere, from emission to deposition, is depicted in Figure 1.1. As mentioned above, primary emissions from various sources are complemented by secondary aerosol production in the atmosphere. Secondary (number and/or mass) formation occurs when precursor gases are oxidized to low-volatility products, and these then nucleate to new particles, or condense onto pre-existing particles. Coagulation and chemical reactions on the particle surface contribute to the “aging” of aerosol particles. “Dry deposition” includes two processes: diffusional deposition to surfaces and sedimentation. Hygroscopic aerosols, like salts, tend to take up water and swell, even if the relative humidity is below 100%. Above water saturation, aerosol particles can pass a threshold radius determined by their composition and initial size and then grow rapidly (“activate”) to cloud droplets, which are several micrometers large. Cloud droplets can collect more interstitial particles via diffusion and inertial impaction. These two mechanisms of aerosol mass and number transfer into the condensed cloud phase are often referred to as “nucleation scavenging” and “collision scavenging”, respectively. Atmospheric gases can also dissolve in cloud droplets and undergo chemical reactions in the aqueous phase, which can result in products with a low volatility. These add to the aerosol mass. Cloud droplets collide with each other and grow, accumulating both dissolved and insoluble aerosol material. When the droplets become large enough to form precipitation and fall out, this is an efficient removal process for the scavenged aerosol (also referred to as “in-cloud scavenging”). More particles can be collected by the sedimenting precipitation particles and are removed from the atmosphere by “below-cloud scavenging”. In-cloud and below-cloud scavenging together are called “wet deposition”. However, many hydrometeors evaporate before reaching the ground, and release the collected aerosol material as one particle back into the atmosphere. This particle has grown compared to the initial cloud condensation nucleus by collisions with other particles and/or chemical reactions and can therefore be activated more readily in a subsequent cloud cycle (e.g. Wurzler et al., 2000).

1.2 Aerosol indirect effects on climate via clouds

Aerosol particle number concentrations vary typically from about 20 cm$^{-3}$ in clean polar air to more than 100000 cm$^{-3}$ under polluted urban conditions (Seinfeld and Pandis, 1998). In general, concentrations are lower over the oceans than over land. Human activity has led to a drastic increase in aerosol number and mass concentrations, and a change in aerosol composition. Additionally to the direct interaction with radiation (through scattering and absorption of shortwave and longwave radiation, “aerosol
direct effect”), anthropogenic aerosols have an influence on clouds. Lohmann and Feichter (2005) give an overview over the aerosol indirect effects on clouds, which are comparable in magnitude to the direct effect, but highly uncertain. Aerosol direct and indirect effects are often quantified as “radiative forcings”, a concept originally developed for long-lived greenhouse gases. The IPCC (Intergovernmental Panel on Climate Change) reports define radiative forcing as “the change in net (down minus up) irradiance (solar plus longwave; in W m\(^{-2}\)) at the tropopause after allowing for stratospheric temperatures to readjust to radiative equilibrium, but with surface and tropospheric temperatures and state held fixed at the unperturbed values” (Forster et al., 2007). This strict definition precludes aerosol effects on clouds which influence the hydrological cycle and the distribution of water in the troposphere, and also effects of absorbing aerosols which alter the tropospheric temperature distribution. It only includes aerosol direct effects and the cloud albedo effect (see below) as radiative forcings, while all other interactions - although they can influence the climate system substantially - are considered as feedbacks. Nevertheless, concepts have been developed to quantify the net perturbation of the radiative budget at the top of the atmosphere by aerosol-climate interactions which require changes in the tropospheric state before the perturbation can be calculated. The most common approach is to calculate a fixed sea surface temperature forcing (Lohmann and Feichter, 2005), which is in practice deduced from the difference between two simulations with aerosol emissions representative for present-day and preindustrial conditions, respectively, but with the same prescribed sea surface temperatures. As stratospheric adjustment has little effect on the radiation imbalance, top-of-the-atmosphere rather than tropopause values are used for simplicity.

The level of scientific understanding for anthropogenic aerosol effects on clouds is classified as “low” for the cloud albedo effect (explained below), but “very low” for all other aerosol indirect effects, by the IPCC (Denman et al., 2007).

### 1.2 Aerosol indirect effects on climate via clouds

#### 1.2.1 Aerosol indirect effects via liquid clouds

Aerosol effects on liquid clouds have been studied for more than 20 years with models on different scales. Some observational support is available, but the global effect can only be inferred from climate model simulations.

**Cloud albedo effect**

Because a subset of the atmospheric aerosol particles acts as cloud condensation nuclei, increased aerosol concentrations can increase the number of cloud droplets in clouds. At a fixed cloud liquid water content, this reduces the mean droplet size and increases the cloud albedo (Twomey, 1974), if the absorption by the aerosols themselves is low or moderate. This effect has been termed “Twomey effect”, “first indirect effect”, or “cloud albedo effect” and is considered as a radiative forcing by the IPCC (Forster et al., 2007), because it comprises only changes of the radiative properties of the clouds. Observational evidence of the cloud albedo effect is provided by in-situ measurements of cloud microphysical properties in ship-tracks, e. g. by Ferek et al. (1998) who report increased droplet concentrations and reduced droplet effective radii at a relatively constant liquid water content.

The radiative forcing by the cloud albedo effect due to anthropogenic aerosols is estimated by global models to range between \(-0.3\) to \(-1.8\) W m\(^{-2}\), with a best estimate of \(-0.7\) W m\(^{-2}\) (Forster et al., 2007).

**Cloud lifetime effect**

Increased droplet number and reduced droplet size influences the rate of droplet collision-coalescence and slows down precipitation formation. Baker and Charlson (1990) show that two stable regimes of low and high cloud condensation nuclei (CCN) concentrations exist in the cloud-topped boundary layer. While the low-CCN regime is maintained by efficient precipitation formation and aerosol washout, the precipitation sink is inactive for high CCN concentrations. Albrecht (1989) hypothesized that drizzle reduction due to increased aerosol concentrations in shallow marine clouds could increase the cloud lifetime and low-level cloudiness. In the literature this has been referred to as “Albrecht effect”, “second indirect effect” or “cloud lifetime effect” (Forster et al., 2007). While suppression of drizzle has been observed in ship-tracks in several studies (Ferek et al., 2000; Lu et al., 2007), the cloud liquid water content in the polluted regime is sometimes higher, sometimes lower than under comparable clean conditions. An observed aerosol impact on cloud lifetime has not been reported yet. While most global climate models do simulate an increase in cloud lifetime in accordance with Albrecht’s (1989) hypothesis (Lohmann and Feichter, 2005), large-eddy simulations suggest that the lifetime of polluted clouds can also be reduced due to enhanced evaporation of small droplets (Jiang et al., 2006).
Global climate model estimates of the cloud lifetime effect due to anthropogenic aerosols, which is difficult to distangulate from the cloud albedo effect, range between $-0.3$ to $-1.4$ W m$^{-2}$ for the change in top-of-the-atmosphere net radiation (Lohmann and Feichter, 2005).

1.2.2 Aerosol indirect effects via mixed-phase clouds

Mixed-phase clouds, containing both liquid and frozen water, are frequent in mid-latitude and polar regions (Quante, 2004). About 70% of the global precipitation forms via the ice phase (Lau and Wu, 2003). At temperatures below 0°C, water droplets do not freeze spontaneously, but can persist as supercooled liquid until approximately $-38^\circ$C, the homogeneous freezing temperature. At warmer temperatures, freezing of droplets occurs only through heterogeneous nucleation on ice nuclei. Ice nuclei are predominantly insoluble particles, such as mineral dust or soot (Richardson et al., 2007), but what exactly is required for a particle to be a good ice nucleus is unknown. Freezing occurs via different mechanisms (deposition, immersion, condensation and contact freezing). The difference and relative importance of these nucleation modes is difficult to observe in the atmosphere. In the presence of unfrozen droplets, the air is saturated with respect to supercooled liquid water, and thus it is supersaturated with respect to ice. This imbalance leads to rapid growth of the ice crystals with subsequent evaporation of liquid droplets once the relative humidity drops below water saturation, the so-called Wegener-Bergeron-Findeisen process (Findeisen, 1938). Therefore precipitation formation is more efficient in mixed-phase clouds than in warm clouds (Rogers and Yau, 1989). Aerosol indirect effects on mixed-phase cloud processes have only been investigated in recent years. So far, no quantitative global estimates of the change in top-of-the-atmosphere net radiation by aerosol indirect effects on mixed-phase clouds have been given. Our study in chapter 5 (Hoose et al., 2008a) finds a small offset to the aerosol indirect effect on liquid clouds of $+0.1$ W m$^{-2}$ in the top-of-the-atmosphere shortwave radiation on the Northern Hemisphere, caused by two of the below-mentioned processes: the glaciation indirect effect and the deactivation indirect effect.

Thermodynamic indirect effect in deep convective clouds

In deep convective clouds, freezing is usually thought to be initiated heterogeneously at some point once the cloud droplets are sufficiently supercooled. Rosenfeld and Woodley (2000) measured supercooled liquid cloud droplets at the top of continental deep convective clouds at temperatures down to $-37.5^\circ$C, the homogeneous freezing temperature. Khain et al. (2001) demonstrated that these observations could only be reproduced in a numerical cloud model if the droplet concentrations were sufficiently high and the individual droplets hence small. Small droplets have a low collision efficiency and a low immersion freezing probability. In a subsequent modeling study (Khain et al., 2005), it was shown that the suppression of warm rain formation and the delayed freezing in polluted clouds result in increased latent heat release by freezing at higher altitudes, invigoration of convection, cloud extent to higher altitudes and triggering of secondary clouds in the vicinity. Koren et al. (2005) confirmed the model results by statistical analysis of satellite data over the North Atlantic Ocean, suggesting systematic invigoration of convective clouds by pollution, desert dust and biomass burning aerosols. In the IPCC report (Denman et al., 2007), this effect is termed "thermodynamic effect" because the anthropogenic aerosols are assumed to influence the thermodynamic structure of convective clouds.

Glaciation indirect effect

Lohmann (2002a) proposed that anthropogenic soot aerosols, shown to have a certain ice nucleating ability in laboratory experiments (e.g. Diehl and Mitra, 1998), can increase glaciation of stratiform supercooled liquid clouds. This entails a higher precipitation efficiency and shorter cloud lifetime, counteracting the aerosol indirect effects on warm clouds. Lohmann and Diehl (2006) and Storelvmo et al. (2008) showed that the magnitude of this so-called "glaciation indirect effect" depends strongly on assumptions on the efficiency of the natural dust ice nuclei. In chapter 5, this is investigated in further detail.

Riming indirect effect

Ice and snow crystals in mixed-phase clouds grow by diffusion of water vapor from evaporating droplets and, once they are large enough to sediment, by riming, i.e. collisions with supercooled droplets which freeze onto the crystal surface. The riming efficiency decreases for smaller droplets (Pruppacher and Klett, 1997). Borys et al. (2003) observed a reduction of both riming and snowfall rate in polluted clouds with more numerous but smaller droplets. In global simulations, this "riming indirect effect" was not
confirmed to decrease the snowfall rate on a global scale, because of feedbacks of the aerosol indirect effects on liquid clouds on the climate system (Lohmann, 2004).

Deactivation indirect effect

As mentioned above, ice nucleation operates in different modes. It can be assumed that immersion and contact nucleation are the dominant freezing processes in mixed-phase clouds, because they involve the preexisting liquid droplets. For contact freezing, a dry interstitial aerosol particle collides with a supercooled droplet and initiates freezing, while for immersion freezing, a particle contained inside a liquid droplet (which it has entered at a warmer temperature by either nucleation or collision scavenging) causes the freezing. For a given ice nucleating material, immersion freezing generally occurs at lower temperatures than contact freezing (e.g. Pitter and Pruppacher, 1973). It has been suggested that heterogeneous freezing is initiated by “active sites” at the aerosol surface (Pruppacher and Klett, 1997). As these sites are possibly vulnerable to partial dissolution when an ice nucleus is immersed in a droplet (Fletcher, 1970), this can explain the higher freezing efficiency in the contact mode when the particle surface is dry.

Coating with a hygroscopic material, like anthropogenic sulfate, could inhibit particles to act in the contact mode. Thus the freezing temperature would shift to the lower immersion freezing threshold, an apparent deactivation, as freezing at a given temperature is a stochastical process. Reduction in ice nuclei concentrations connected to human pollution have been reported during the passage over a big city (Braham and Spyers-Duran, 1974) and during Arctic haze conditions (Borys, 1989). Laboratory experiments have shown that soot coated with sulfate nucleates ice less efficiently in the deposition mode than uncoated soot (Möhler et al., 2005). Girard et al. (2005) have parameterized ice nuclei deactivation as a function of sulfate concentration in a single column model simulation of Arctic clouds. Storelvmo et al. (2008) calculate the partitioning of mineral dust into potential contact and immersion nuclei based on sulfate concentrations in a global climate model. In chapter 5, we explore the deactivation indirect effect in the global aerosol-climate model ECHAM5-HAM.

1.3 Aerosol-cloud interactions in global climate models

1.3.1 Global aerosol modeling

Because of computational constraints, global climate (or general circulation) models (GCMs) have to represent the aerosol composition, size distribution and life cycle in a simplified way. Therefore GCMs which compute aerosol-cloud interactions online account only for a selection of aerosol chemical components and a simplified representation of the size distribution. Chemical transport models (CTMs), which use
OFFLINE METEOROLOGICAL FIELDS, CAN AIM FOR A BETTER, COMPUTATIONALLY MORE EXPENSIVE REPRESENTATION WITH MORE SPECIES AND/OR A HIGHER RESOLUTION OF THE AEROSOL SIZE DISTRIBUTION.

FIGURE 1.2 GIVES AN OVERVIEW OF AEROSOL EMISSIONS AND BURDENS IN STATE-OF-THE-ART GCMs AND CTMs. THESE RESULTS FROM THE AEROCom INTERCOMPARISON PROJECT REVEAL LARGE INTERMODEL DIFFERENCES. THE TOTAL AEROSOL EMISSIONS VARY BY MORE THAN AN ORDER OF MAGNITUDE, WITH AN ALL-MODELS AVERAGE OF 18800 Tg yr$^{-1}$. SOURCES ARE DOMINATED BY SEA SALT WITH 16600 Tg yr$^{-1}$, FOLLOWED BY DUST (1840 Tg yr$^{-1}$), SULFATE (179 Tg yr$^{-1}$), PARTICULATE ORGANIC MATTER (96.6 Tg yr$^{-1}$), AND FINALLY BLACK CARBON (11.9 Tg yr$^{-1}$) (Textor et al., 2006). THE SULFATE SOURCE TERM INCLUDES BOTH PRIMARY AND SECONDARY SULFATE FORMATION. THE INTERMODEL DIVERSITY IN THE SEA SALT AND DUST EMISSIONS IS PARTLY DUE TO WHETHER SUPERCOARSE PARTICLES, WHICH HAVE SHORT ATMOSPHERIC LIFETIME, ARE CONSIDERED OR NOT. THE RESULTING DIVERSITY IN THE AEROSOL BURDENS IS SMALLER THAN IN THE EMISSIONS. IN MOST MODELS, THE TOTAL AEROSOL BURDEN IS DOMINATED BY MINERAL DUST (ON AVERAGE 19.2 Tg), FOLLOWED BY SEA SALT WITH 7.5 Tg, ALTHOUGH SEA SALT EMISSIONS ARE HIGHER BY AN ORDER OF MAGNITUDE. THIS INDICATES MORE EFFICIENT REMOVAL PROCESSES FOR SEA SALT THAN FOR MINERAL DUST. SEA SALT IS EMMITED IN AN ENVIRONMENT FAVORABLE FOR EFFICIENT WET SCAVENGING (FREQUENT CLOUD AND PRECIPITATION OCCURRENCE OVER THE OCEANS), WHILE MINERAL DUST IS ASSOCIATED WITH DRY AIR MASSES. ALSO, UNCOATED MINERAL DUST AEROSOLS ARE LESS EFFICIENT CLOUD CONDENSATION NUCLEI THAN THE HYGROSCOPIC SALT PARTICLES. THE BURDENS OF SULFATE AND PARTICULATE ORGANIC MATTER ARE COMPARABLE WITH 2 Tg AND 1.7 Tg. AS THE EMISSIONS OF THE ORGANIC MATTER ARE ONLY HALF OF THE SULFATE EMISSIONS, THIS SUGGESTS MORE EFFICIENT REMOVAL PROCESSES FOR SULFATE THAN FOR PARTICULATE ORGANIC MATTER (SIMILAR TO WHAT HAS BEEN DISCUSSED FOR SEA SALT VERSUS MINERAL DUST ABOVE). THE BLACK CARBON BURDEN IS ON AVERAGE 0.24 Tg. FIGURE 1.2 ILLUSTRATES THAT UNCERTAINTIES REMAIN BOTH REGARDING THE EMISSION AND THE ATMOSPHERIC FATE OF AEROSOL PARTICLES, ESPECIALLY BECAUSE OF LACK OF OBSERVATIONAL CONSTRAINTS.

1.3.2 Aerosol-cloud coupling

Modeling aerosol indirect effects in GCMs is usually restricted to stratiform clouds, because convective clouds are assumed to exist only on timescales comparable to or smaller than the integration timestep. The representation of cloud droplet number is required, in addition to the cloud water mass. Earlier studies related cloud droplet number empirically to sulfate aerosol mass (Boucher and Lohmann, 1995, and others), using the resulting effective droplet radius for calculation of the cloud radiative properties and the cloud albedo effect. When, in addition to the cloud radiative properties, the precipitation rate is calculated as a function of droplet number (Lohmann and Feichter, 1997; Rotstayn, 1999; Kristjánsson, 2002; Menon et al., 2002), the cloud lifetime effect can also be simulated. In a more physical approach, prognostic equations for cloud droplet number (a so-called “double-moment scheme”) are included in a number of state-of-the-art GCMs (Ghan et al., 1997; Lohmann et al., 1999; Storelvmo et al., 2006, and others). With the prognostic treatment, transport, nucleation, collision-coalescence, freezing, melting and evaporation of cloud droplets can be taken into account and influence the cloud properties at subsequent timesteps.

Continuity equations for cloud droplet concentrations ($N_l$) have the following form:

$$\frac{dN_l}{dt} = \text{Transport} + \text{Nucleation} + \text{Melting} - \text{Freezing} - \text{Evaporation}$$

$$- \text{Accretion by rain and snow} - \text{Autoconversion} - \text{Selfcollection} \quad (1.1)$$

Transport includes advective, turbulent and convective contributions. Nucleation is the formation of cloud droplets on aerosol particles; hence, this term is directly coupled to the aerosol concentration and composition. Melting of ice crystals is a process of second-order importance and is neglected in some models. The terms in the second line all refer to collision processes, and the distinction is somewhat artificial. Precipitating rain and snow particles are usually not treated prognostically, because they are assumed to fall out relatively fast. Cloud droplets colliding with them add to the precipitating water mass. Autoconversion and selfcollection both refer to collisions among cloud droplets. The droplets resulting from an autoconversion process are assumed to be big enough to fall out from the cloud, while droplets resulting from a selfcollection process remain in the cloud, increasing the mean droplet size. As the collision efficiency is a function of droplet size, these terms are implicitly influenced by aerosol concentrations.

Figure 1.3 gives an example of a cloud droplet nucleation parameterization. Lin and Leaitch (1997) give the number of droplets as a function of available aerosol particles larger than a certain cut-off diameter and vertical velocity. The parameterization was obtained by fitting measured droplet and aerosol concentrations from a marine environment. The more aerosol particles are available, and the
higher the vertical velocity, the more droplets form. The fraction of activated particles increases with increasing vertical velocity, but does not reach values higher than 80% even for a vertical velocity of 5 m s\(^{-1}\). In other parameterizations (e. g. Ghan et al., 1993), all aerosol particles are assumed to activate if their number is low.

Two examples of parameterizations of the autoconversion process are displayed in Figure 1.4. For a given cloud water content, the conversion of cloud water to rain water is slower, the higher the droplet concentration – because higher droplet concentrations signify smaller mean droplet sizes and lower collision efficiencies, and on average more collisions are required to form a precipitation-size droplet. One of the parameterizations has been derived from theoretical considerations (Beheng, 1994), and the other one from large-eddy simulations with a detailed representation of the droplet size distribution (Khairoutdinov and Kogan, 2000). Both agree reasonably well with explicitly calculated autoconversion rates over a wide range of conditions in stratiform boundary layer clouds (Wood, 2005b). The parameterization by Beheng (1994) is more sensitive to the cloud droplet number, and therefore presumably results in a larger cloud lifetime effect. In how far these parameterizations, which have been developed only for a limited range of parameters and conditions, can be applied to global models, is unclear. In GCMs, the autoconversion rate is usually tuned, i. e. scaled with a constant parameter, in order to achieve balance of the in- and outgoing radiation at the top of the atmosphere.
Freezing of droplets can occur homogeneously (below $-38^\circ$C) or heterogeneously. Heterogeneous freezing is thought to depend on ice nuclei concentrations, and therefore on aerosol properties, but the dependency is difficult to parameterize. Often simple temperature-dependent parameterizations are used instead, but for simulation of the glaciation indirect effect (Lohmann, 2002a; Storelvmo et al., 2008, and chapter 5), the freezing term has to be linked to aerosol parameters. Commonly used ice nuclei concentration parameterizations as shown in Figure 1.5 are steep functions of temperature (or, alternatively, supersaturation). They disagree by orders of magnitude, illustrating the uncertainties connected to modeling heterogeneous freezing. Gultepe et al. (2001) demonstrated that average cloud ice crystal concentrations do not correlate well with temperature, and ice nuclei parameterizations match observed ice crystal concentrations only in a small range of temperatures around $\approx -20^\circ$C. In some models (e.g. Meyers et al., 1992), the ice nuclei concentrations obtained by a temperature- or supersaturation-dependent parameterization are directly translated into ice crystal concentrations. Some of the original formulations (e.g. Young, 1974a) additionally take into account the collision probability between ice nuclei and droplets, which depends on particle and droplet sizes. The number of frozen droplets is in this case determined by the collision rate, the integration time and the original droplet number. In Figure 1.5, the number of frozen droplets, calculated by Young’s (1974a) parameterization for Brownian diffusion, is included for comparison. Depending on the chosen parameters (particle sizes, droplet concentration, integration time) it can vary over several orders of magnitude.

In most GCMs, cloud effects on aerosols are reduced to two processes: in-cloud sulfate production and wet deposition. In-cloud sulfate production accounts for more than half of the global airborne sulfate (Barth et al., 2000). Several reactions can result in the formation of sulfate in the liquid phase. Oxidation of dissolved SO$_2$ by O$_3$ and H$_2$O$_2$ are most commonly considered in models, requiring simplifying assumptions about the pH-value of the cloud water (Feichter et al., 1996; Roelofs et al., 2006).

In-cloud wet deposition is most commonly handled with prescribed scavenging ratios (Textor et al., 2006), i.e. the fraction of aerosol incorporated into cloud hydrometeors and removed proportionally to the precipitation of cloud mass is set to a fixed value for each component. These scavenging ratios decrease with solubility and are usually highest (often 1) for sea salt and lowest (often 0) for insoluble black carbon. Scavenging into the ice phase is in some models assumed to be smaller than scavenging into the liquid phase, because a large fraction of the droplets evaporates during cloud glaciation (Wegener-Bergeron-Findeisen-process) and releases the formerly scavenged aerosol particles (Verheggen et al., 2007).

Below-cloud scavenging is parameterized as a function of the rain/snow rate. Aerosol models are very sensitive on the treatment of wet deposition, because it is the dominant removal process for anthropogenic aerosol components (sulfate, black carbon, particulate organic matter) and contributes to roughly half of the deposition for dust and sea salt (Textor et al., 2006).

### 1.3.3 ECHAM5-HAM

For the studies presented in this dissertation, the global climate model ECHAM5 (Roeckner et al., 2003) is used. It is the 5th generation of the global climate model ECHAM, derived from the ECMWF (European
1.3 Aerosol-cloud interactions in global climate models

Centre for Medium-Range Weather Forecasts) model, and developed at the Max-Planck Institute for Meteorology, Hamburg. The stratiform cloud scheme contains double-moment representations for both the liquid and the ice phase (Lohmann et al., 1999; Lohmann, 2002b; Lohmann et al., 2007).

In the aerosol module HAM (labelled MPIHAM in Figure 1.2), the aerosol population is described by the superposition of 7 lognormal modes (Stier et al., 2005; Vignati et al., 2004). 5 components (sulfate, black carbon, particulate organic matter, sea salt and dust) are considered. The standard deviations of the modes are prescribed, and the prognostic variables are the component masses and the number concentrations for the 7 modes (some of which represent internal mixtures of different components, e.g. mineral dust coated with sulfate). Emissions of anthropogenic pollutants and from wildfires and vegetation are prescribed, while sea salt and dust emissions are calculated interactively depending on the modelled surface wind.

1.3.4 Mixed-phase clouds in ECHAM5-HAM

The representation of stratiform mixed-phase clouds in ECHAM5-HAM is determined by the parameterization of two processes: heterogeneous freezing and the Wegener-Bergeron-Findeisen process. Heterogeneous freezing is calculated depending on temperature and on the concentration of the possible ice nuclei black carbon and mineral dust, as described in Lohmann and Diehl (2006) and chapter 5. When the ice mass, formed by heterogeneous freezing or ice multiplication, or fallen into a liquid cloud layer from above, exceeds a threshold value of 0.5 mg kg$^{-1}$, the remaining cloud droplets are assumed to evaporate and the excess water vapor is deposited to the ice crystals within one model timestep (Lohmann et al., 2007). Below –35°C, all remaining droplets are frozen, regardless of ice nuclei concentrations (homogeneous freezing).

Figures 1.6 and 1.7 show that, in a statistical sense, the liquid and ice water content in stratiform clouds in ECHAM5-HAM is in good agreement with aircraft observations. The label ECHAM5-RH refers to the use of a cloud cover scheme depending only on relative humidity (Sundqvist et al., 1989), which was also employed in the global simulations in chapters 3 and 5. Figure 1.6 illustrates that the simulated average in-cloud liquid water content decreases with temperature below the freezing point, from 0.12 g m$^{-3}$ at 0°C to 0.01 g m$^{-3}$ below –35°C. The simulated ice water content is rather constant (around 0.02 g m$^{-3}$) over this temperature range, with a drop only below –35°C to 0.01 g m$^3$. This behavior is similar to observations, although the observed liquid water content is lower than simulated and falls below the average ice water content around –20°C. In Figure 1.7 the partitioning between ice and liquid is examined. ECHAM5-HAM in general simulates either low (<0.1) or high (>0.9) ice mass fractions, and the probability of low ice mass fractions decreases with temperature, while the probability of high ice mass fractions increases accordingly. Observations report a similar picture, except that ice mass fractions between 0.7 and 0.9 have a higher occurrence probability than simulated.
1 Introduction: Aerosol effects on clouds and cloud effects on aerosols

Figure 1.7: Probability distributions of mixed-phase cloud composition (ice water content (IWC) / total (liquid plus ice) water content (TWC)) for different temperature intervals from ECHAM5-HAM simulations as compared to observations by Korolev et al. (2003) (Lohmann et al., 2007).

1.4 Overview over this dissertation

This dissertation includes several studies on the simulation of aerosol-cloud interactions in ECHAM5-HAM. The focus is on mixed-phase clouds, a field which has received limited interest so far, although clouds containing both liquid and ice are frequent over wide ranges of the globe. Chapters 2 and 3 treat aerosol processing in clouds. A novel treatment of in-droplet and in-crystal particles is introduced. In a single-column model study, the performance of the new scheme in ECHAM5-HAM can be evaluated in comparison to aerosol size distribution measurements in different mixed-phase clouds (Hoose et al., 2008b). A global view of aerosol processing in liquid, mixed-phase and ice clouds is presented in chapter 3. Chapter 4 discusses mixed-phase clouds in a special environment, the Arctic, and presents single-column simulations which were conducted as part of a model intercomparison study (Klein et al., 2007). Aerosol indirect effects on mixed-phase clouds, inferred from global simulations, are the subject of chapter 5 (Hoose et al., 2008a). Model assumptions about the freezing efficiency of natural mineral dust particles are compared, and the competition between the glaciation indirect effect and the deactivation indirect effect is evaluated.

The summary and outlook in chapter 6 provide perspectives how the explicit representation of in-cloud particles can be used for a more physical representation of heterogeneous ice nucleation. This will finally link the two big subjects of this study: in-cloud processes influencing aerosol particles and aerosol particles influencing the evolution of mixed-phase clouds through ice nucleation.
2 Aerosol processing in mixed-phase clouds in ECHAM5-HAM: Model description and comparison to observations

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Abstract

The global aerosol-climate model ECHAM5-HAM has been extended by an explicit treatment of cloud-borne particles. Two additional modes for in-droplet and in-crystal particles are introduced, which are coupled to the number of cloud droplet and ice crystal concentrations simulated by the ECHAM5 double-moment cloud microphysics scheme. Transfer, production and removal of cloud-borne aerosol number and mass by cloud droplet activation, collision scavenging, aqueous-phase sulfate production, freezing, melting, evaporation, sublimation and precipitation formation are taken into account. The model performance is demonstrated and validated with observations of the evolution of total and interstitial aerosol concentrations and size distributions during three different mixed-phase cloud events at the alpine high-altitude research station Jungfraujoch (Switzerland). Although the single-column simulations can not be compared one-to-one with the observations, the governing processes in the evolution of the cloud and aerosol parameters are captured qualitatively well. High scavenged fractions are found during the presence of liquid water, while the release of particles during the Wegener-Bergeron-Findeisen process results in low scavenged fractions after cloud glaciation. The observed coexistence of liquid and ice, which might be related to cloud heterogeneity at subgrid scales, can only be simulated in the model when assuming non-equilibrium conditions.

2.1 Introduction

Cloud processes have important effects on atmospheric aerosol concentration, size distribution, chemical composition and mixing state. Pruppacher and Jaenicke (1995) estimated that on average an aerosol particle sampled in a remote location has undergone three cloud cycles. This involves the following processes: Aerosol particles, preferably the bigger and hygroscopic ones, act as the cloud condensation nuclei on which cloud droplets form. Cloud droplets can collect more aerosol particles and other cloud droplets by collisions. The soluble part of the aerosol particles dissolves in the water. Additionally, atmospheric gases can also transfer into droplets and undergo chemical reactions in the aqueous phase. If precipitation is formed, all material collected in the droplets is removed (scavenged) from the atmosphere. However, a large fraction of clouds does not form precipitation, but evaporates (Lin and Rossow, 1996). In this case the dissolved material concentrates in the liquid phase again or crystallizes, and together with the possible insoluble material contained inside the droplet forms one new, mixed aerosol particle (Pruppacher and Klett, 1997). These reemitted particles are larger than before cloud processing. Hoppel et al. (1986) and Hoppel et al. (1990) have observed bimodal aerosol size distributions in the marine boundary layer. They are explained with the growth of activated particles through cloud processing to form the larger mode, while freshly nucleated, not activated particles constitute the smaller mode. A distinct bimodal shape which is most probably generated and maintained by cloud processes was also found at the high-alpine site Jungfraujoch (Switzerland) (Weingartner et al., 1999).

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Cloud droplets can also undergo freezing. Less is known about processing of aerosol particles in ice crystals. At temperatures above −38°C, an ice nucleus is required to initiate (heterogeneous) freezing. This ice nucleus can either be already immersed in the droplet (immersion freezing, or condensation freezing if the droplet formed on the same particle) or it can be an interstitial particle colliding with a droplet (contact freezing). Ice can also deposit directly from the vapor phase on a dry aerosol particle (deposition nucleation), or droplets can possibly freeze during evaporation. Ice crystals can collide with other hydrometeors (aggregation, riming) or with aerosol particles. Ice crystal multiplication can result from shattering of freezing droplets or fracturing of crystals. Precipitation formation is more efficient than via the liquid or the ice phase alone, because the difference in saturation vapor pressure over ice and over water leads to evaporation of the droplets and rapid growth of the ice crystals, if they coexist in a mixed-phase cloud (Findeisen, 1938). In the following this is called Wegener-Bergeron-Findeisen process. If ice crystals do not form precipitation, they can either melt again or sublimate directly, releasing the aerosol contained in them back to the interstitial air.

Models describe aerosol processing in clouds with different degrees of complexity. With few exceptions, most global aerosol models do not treat in-droplet aerosols explicitly, but account for effects of cloud processing in simplified ways. Sulfate formation in cloud droplets is estimated to contribute to up to 80% of the total global sulfate production (Barth et al., 2000). It is therefore taken into account in many global and regional aerosol models via addition of sulfate to the accumulation and/or coarse mode (e.g. Barth et al., 2000; Adams and Seinfeld, 2002; Liu et al., 2005; Stier et al., 2005; Gong et al., 2006; Roelofs et al., 2006). Size-resolved cloud chemical models calculate 2-3 times higher sulfate production rates than bulk models, due to the variability of cloud water pH among cloud droplets of different sizes (Kreidenweis et al., 2003). However, global models can in general not account for different chemical compositions of cloud droplets of different sizes (Adams and Seinfeld, 2002; Stier et al., 2005). Roelofs et al. (2006) represented this effect in a simplified way in ECHAM5-HAM by assuming two bins of concentrated and diluted droplets.

The only global aerosol-climate models that treat cloud-borne particles explicitly as independent tracers are the Hadley Center climate model (HADAM4), the Goddard Institute for Space Studies (GISS) ModelE and the Model for Integrated Research on Atmospheric Global Exchanges (MIRAGE) modeling system. HADAM4 (Jones et al., 2001) calculates sulfate mass for three modes, one of which represents sulfate dissolved in cloud droplets. In GISS ModelE (Koch et al., 2006), a stratiform dissolved species budget is calculated for all soluble and partially soluble aerosol and gas-phase species, saving the dissolved fraction for the duration of the cloud lifetime. As GISS ModelE simulates only the mass and not the number of aerosols, the influence of cloud processing on aerosol size is not treated. MIRAGE (Easter et al., 2004) explicitly predicts mass and number of cloud-borne particles in four different lognormal modes and for six different chemical components. In HADAM4 and MIRAGE, transfer into these modes is possible via activation and, for Aitken mode particles only, Brownian diffusion. Ghan and Easter (2006) investigate how this treatment in MIRAGE can be simplified. They conclude that neglecting transport and predicting the total mass of cloud-borne aerosol particles instead of different chemical components separately introduces little error.

Models operating on smaller scales can treat cloud-borne aerosol particles with a higher degree of complexity. Mechem et al. (2006) and Ivanova and Leighton (2008) investigate cloud processing via activation and collision-coalescence of cloud droplets in mesoscale models. Yin et al. (2005) study the relative importance of nucleation scavenging, impaction scavenging and in-cloud chemistry for budgets of cloud-borne sulfate aerosols in a mixed-phase convective cloud with a bin-resolved cloud model. They find that nucleation scavenging dominates and impaction scavenging plays only a minor role for the mass budget. Aqueous-phase chemistry accounted for < 17% of the in-cloud sulfate mass in the simulated case. However, it can be expected that the relative importance of the different processes depends on the precursor concentrations and on the cloud type. Romakkaniemi et al. (2006) investigate the influence of nitric acid on cloud processing, using a parcel model with a moving sectional representation of the aerosol size distribution. They account for liquid-phase thermodynamics, which influences the sulfate production, and coagulation.

In contrast to HADAM4 and MIRAGE, the treatment of aerosol processing in this study considers liquid and ice hydrometeors separately. This allows taking processes in mixed-phase clouds into account. Especially heterogeneous freezing and the Wegener-Bergeron-Findeisen process is expected to influence aerosol processing. The version of ECHAM5-HAM used in this study (Lohmann et al., 2007) couples aerosol and cloud microphysics (via activation of cloud droplets, heterogeneous freezing, and cloud processing of aerosol particles), allowing feedbacks in both ways: Cloud droplet number concentrations depend on the number and size of the available aerosol particles, and heterogeneous freezing is
2.2 Model description

2.2.1 ECHAM5

For this study, we use the coupled aerosol-climate model ECHAM5-HAM (Stier et al., 2005) with a microphysics module that includes prognostic equations for cloud droplet number concentration (Lohmann et al., 1999) and ice crystal number concentration (Lohmann, 2002b; Lohmann and Diehl, 2006). ECHAM5 is the fifth version of the global climate model developed by the Max Planck Institute in Hamburg, which is derived from the global model of the European Center for Medium-range Weather Forecasts. HAM stands for Hamburg Aerosol Module. In the model version used here, cloud droplet and ice crystal concentrations depend on the available aerosol (Lohmann et al., 2007). Cloud cover is calculated by a statistical approach (Tompkins, 2002). ECHAM5-HAM can be set up as a single-column model (SCM), with exactly the same code for the model physics as the global model. Here we run the model in single-column mode on 31 vertical levels with a timestep of 15 minutes.

2.2.2 Introducing cloud-borne modes in ECHAM5-HAM

The aerosol module HAM (Stier et al., 2005) describes the atmospheric aerosol population as a superposition of seven lognormally distributed modes. As their standard deviations are prescribed, only the total number concentration and the masses of the different chemical components (sulfate, black carbon, organic carbon, sea salt and mineral dust) are prognostic variables in the model. The modes are divided into soluble, internally mixed modes (containing sulfate) and insoluble, externally mixed modes, which are assigned to different size ranges. The actual modal diameters can vary and are calculated from the mass and number for each mode. Figure 2.1 lists the seven modes of the standard ECHAM5-HAM, their size ranges, components and acronyms.

In the standard version of HAM, these seven modes describe the total (in-cloud plus interstitial) aerosol. For the calculation of wet deposition, the partitioning between in-cloud and interstitial is prescribed in a crude way using fixed scavenging ratios, defined as the fraction of particle mass and number that is embedded in the cloud water. These scavenging ratios depend only on the mode, temperature (as an indicator for liquid/mixed-phase/ice clouds) and on whether the clouds are convective or stratiform.

In the version described in this article, we have introduced two new modes which explicitly represent cloud-borne particles: one in-droplet mode (acronym CD for cloud droplet) and one in-crystal mode (acronym IC for ice crystal). The seven standard modes then only contain interstitial aerosol, as all material which is taken up into cloud droplets or crystals is transferred into the two new modes CD and IC. Both cloud-borne modes can contain all five chemical components present in the standard modes. They are not assigned to any specified size range, as their size is calculated as described below.

The number concentrations of particles in the cloud-borne modes are directly coupled to cloud microphysics. It is assumed that the number of particles in cloud droplets is equivalent to the number of cloud droplets and that the number of particles in ice crystals is equivalent to the number of ice crystals. Cloud droplet number \(N_l\) and ice crystal number \(N_i\) are prognostic variables in ECHAM5 (Lohmann et al., 1999; Lohmann, 2002b):

\[
\frac{dN_l}{dt} = \left. \frac{dN_l}{dt} \right|_{\text{transp.}} + Q_{\text{nuc}} + Q_{\text{mel}} - (Q_{\text{aut}} + Q_{\text{self}} + Q_{\text{acc}} + Q_{\text{frz}} + Q_{\text{evap}}) \tag{2.1}
\]

\[
\frac{dN_i}{dt} = \left. \frac{dN_i}{dt} \right|_{\text{transp.}} + Q_{\text{nci}} + Q_{\text{frz}} + Q_{\text{secp}} - (Q_{\text{agg}} + Q_{\text{selfi}} + Q_{\text{acci}} + Q_{\text{melt}} + Q_{\text{sub}}) \tag{2.2}
\]

The concentrations \(N_l\) and \(N_i\) are in-cloud values. The first terms on the right-hand sides include all advective, convective and diffusive transport processes. \(Q_{\text{nuc}}\) represents the rate of change in cloud droplet
Figure 2.1: Transfers between the interstitial (standard HAM) modes NS (nucleation soluble), KS (Aitken soluble), AS (accumulation soluble), CS (coarse soluble), KI (Aitken insoluble), AI (accumulation insoluble) and CI (coarse insoluble) and the cloud-borne modes CD (in cloud droplets) and IC (in ice crystals). The number median radius $\bar{r}$ of the lognormal distribution is confined to the given ranges. Chemical components: SO4=sulfate, BC=black carbon, OC=organic carbon, SS=sea salt, DU=dust. Processes affecting aerosol or hydrometeor number or mass: nuci=homogeneous nucleation of ice crystals, colli=collisions between aerosol particles and ice crystals, nuc=nucleation of cloud droplets, coll=collisions between aerosol particles and cloud droplets, frz=freezing, mlt=melting, self=selfcollection of cloud droplets, aqu. chem.=aqueous chemistry, aut=autoconversion of cloud droplets, acc=accretion of cloud droplets by rain or snow, selfi=selfcollection of ice crystals, secp=secondary production of ice crystals, agg=aggregation of ice crystals to snow, acci=accretion of ice crystals by snow, evap=evaporation of cloud droplets, sub=sublimation of ice crystals.
number by nucleation, $Q_{\text{melt}}$ by melting, $Q_{\text{act}}$ by autoconversion to rain droplets, $Q_{\text{self}}$ by selfcollection, $Q_{\text{acc}}$ by accretion by falling rain or snow, $Q_{\text{frz}}$ by heterogeneous freezing in the contact and immersion mode and $Q_{\text{evap}}$ by evaporation during cloud dissipation or by the Wegener-Bergeron-Findeisen process. For processes affecting ice crystal numbers, $Q_{\text{nuc}}$ is the homogeneous nucleation of crystals below −35°C, $Q_{\text{secp}}$ the secondary production via the Hallett-Mossop process, $Q_{\text{agg}}$ the aggregation to snow, $Q_{\text{self}}$ the selfcollection, $Q_{\text{acc}}$ the accretion by falling snow and $Q_{\text{sub}}$ the sublimation. The difference between autoconversion/aggregation and selfcollection is that the resulting particle of a selfcollection event is still a cloud droplet/cloud ice crystal, leaving the cloud water/ice content constant, while by autoconversion/aggregation rain and snow is formed and the cloud water/ice content decreases. Heterogeneous condensation freezing is implicitly included in the immersion freezing term (Lohmann and Diehl, 2006). Deposition nucleation is believed to be of less importance in mixed-phase clouds, where the air is still saturated with respect to water and any of the other three freezing mechanisms would set in at higher temperatures. The parameterizations of all rates are described in detail in Lohmann et al. (1999), Lohmann (2002b) and Hoose et al. (2008a).

These processes affect the aerosol in the interstitial and cloud-borne modes. A schematic depiction of the various interactions is given in Figure 2.1. The interactions can be seen as mass and/or number transfers. The masses $m_x$ of the different components $x$ in the in-droplet (CD) and in-crystal (IC) modes are coupled to these microphysical processes as follows:

\[
\frac{dm_{x,\text{CD}}}{dt} = \frac{dm_{x,\text{CD}}}{dt}_{\text{transport}} + \sum_{j=1}^{4} \frac{m_{x,j}}{N_j} \frac{N_{\text{act},j}}{N_{\text{act},k}} Q_{\text{nuc}} + \frac{m_{x,\text{IC}}}{b N_i} Q_{\text{melt}} - \frac{m_{x,j}}{b N_i} (Q_{\text{frz}} + Q_{\text{evap}}) - \frac{m_{x,\text{CD}}}{q_i} \frac{q_{\text{rain}}}{q_i}
\]

\[
+ \delta_{x,\text{SO4}} \frac{dm_{x,\text{SO4,CD}}}{dt}_{\text{aqueous chemistry}} + \sum_{j=1}^{7} \frac{m_{x,j}}{N_j} Q_{\text{coll},j} (3.2)
\]

\[
\frac{dm_{x,\text{IC}}}{dt} = \frac{dm_{x,\text{IC}}}{dt}_{\text{transport}} + \sum_{j=2}^{4} \frac{m_{x,j}}{N_j} \frac{N_{\text{act},j}}{N_{\text{act},k}} Q_{\text{nuc}} + \frac{m_{x,\text{CD}}}{b N_i} Q_{\text{frz}} - \frac{m_{x,\text{IC}}}{b N_i} (Q_{\text{melt}} + Q_{\text{sub}}) - \frac{m_{x,\text{IC}}}{q_i} \frac{q_{\text{snow}}}{q_i} + \sum_{j=1}^{7} \frac{m_{x,j}}{N_j} Q_{\text{coll},j}
\]

with $x = \text{SO4, BC, OC, SS, DU}$.

$N_i$ is the aerosol number in mode $j$, where $j$ runs from 1 to 7 for the interstitial (standard) modes NS, KS, AS, CS, KI, AI, and CI (see Figure 2.1 for the acronyms). $N_{\text{act},j}$ is the number of particles in mode $j$ activated to cloud droplets, and $N_{\text{act},j}$ the number frozen homogeneously to ice crystals. $\delta_{x,\text{SO4}}$ is 1 for $x=\text{SO4}$ and 0 otherwise. The parameterizations of $N_{\text{act},j}$ and $N_{\text{act},j}$ are explained below. Some of the interstitial modes do not contain all chemical species, e.g. mode AI does not include sulfate and therefore $m_{\text{SO4, AI}} = 0$. $m_{x,\text{CD}}$ and $m_{x,\text{IC}}$ refer to grid-mean values. Thus, the rates $Q$ are scaled with the cloud fraction $b$: $Q = b Q$. In addition, $Q_{\text{nuc}}$, $Q_{\text{nuc}}$, $Q_{\text{evap}}$ and $Q_{\text{sub}}$ account for changes in the cloud fraction:

\[
Q_{\text{nuc}} = b Q_{\text{nuc}} + \max \left( \frac{db}{dt}, 0 \right) N_i
\]

\[
Q_{\text{evap}} = b Q_{\text{evap}} - \min \left( \frac{db}{dt}, 0 \right) N_i
\]

\[
Q_{\text{nuc}} = b Q_{\text{nuc}} + \max \left( \frac{db}{dt}, 0 \right) N_i
\]

\[
Q_{\text{sub}} = b Q_{\text{sub}} - \min \left( \frac{db}{dt}, 0 \right) N_i
\]

Selfcollection ($Q_{\text{self}}$ and $Q_{\text{self}}$) only influences the number of cloud-borne particles, not the total mass. $N_{\text{act},j}$, the number of activated particles in mode $j$, only includes nucleation scavenging, not impaction.
scavenging (which is parameterized separately). Cloud droplet nucleation is calculated following Lin and Leaitch (1997):

$$Q_{\text{nuc}} \equiv \frac{\sum_{k=1}^{4} N_{\text{act},k}}{\Delta t} = \frac{0.1 \times 10^6 \text{m}^{-3}}{\Delta t} \times \left( \frac{w N_{\text{aer}>35 \text{nm}} \times 10^{-6} \text{m}^3}{w + \alpha N_{\text{aer}>35 \text{nm}}} \right)^{1.27}, \quad (2.9)$$

where $w$ is the vertical velocity, $N_{\text{aer}>35 \text{nm}}$ is the number of interstitial particles in the internally mixed modes ($j = 1$ to 4) with wet radii larger than 35 nm, and $\alpha = 2.3 \times 10^{-10} \text{m}^4 \text{s}^{-1}$. $N_{\text{act},j} = 0$ for $j = 5, 6, 7$ (the insoluble modes). This formulation differs from the standard version of ECHAM5-HAM with coupled cloud microphysics (Lohmann et al., 2007), where the number of particles available for activation has to be reduced by the number of cloud droplets at the previous timestep $N_{l,\text{old}}$ to avoid double activation of particles.

$$Q_{\text{nuc,std}} = \frac{0.1 \times 10^6 \text{m}^{-3}}{\Delta t} \times \left( \frac{w (N_{\text{aer}>35 \text{nm}} - N_{l,\text{old}}) \times 10^{-6} \text{m}^3}{w + \alpha (N_{\text{aer}>35 \text{nm}} - N_{l,\text{old}})} \right)^{1.27} \quad (2.10)$$

With the explicit reduction of interstitial aerosol through activation, this becomes redundant.

Ice nucleation below $-35^\circ \text{C}$ is assumed to be homogeneous freezing of supercooled aerosol particles. $N_{\text{act},j}$, the number of aerosols in mode $j$ that freeze to ice crystals, is calculated as a function of the particle size (Kärcher and Lohmann, 2002). $N_{\text{act},j} = 0$ for all modes except for the soluble Aitken, accumulation and coarse modes ($j = 2, 3, 4$). For melting, freezing, evaporation and sublimation, in-cloud mass is changed proportionally to $N_{l}$ and $N_{i}$, respectively. For precipitation formation, the cloud-borne masses are removed proportionally to the ratio of the rain (snow) mass created in one timestep, $q_{\text{rain}}$ ($q_{\text{snow}}$), over the cloud liquid (ice) water mass before precipitation formation, $q_{l}$ ($q_{i}$). For in-droplet mass, an additional source term is the sulfate production in the liquid phase (the sixth term on the right-hand side of equation (2.4)). The sulfate production stems from aqueous-phase oxidation of SO$_2$ by H$_2$O$_2$ and O$_3$. It is calculated in the same way as in the standard version of ECHAM5-HAM (Stier et al., 2005; Feichter et al., 1996). Instead of an explicit treatment, Stier et al. (2005) attributed the sulfate produced in the liquid phase to the soluble accumulation and coarse modes as a crude representation of cloud processing. Here, it is directly attributed to the in-droplet mode. Collisions between aerosol particles and hydrometeors are also considered. $Q_{\text{coll},j}$ is the collision rate of aerosol particles of mode $j$ with liquid droplets and $Q_{\text{coll},i,j}$ the collision rate with ice crystals. These collision rates are calculated from the number concentrations in mode $j$, cloud droplet/ice crystal number concentrations and prescribed collision kernels $K_{i,j}$:

$$Q_{\text{coll},j} = K_{i,j} N_{l} N_{i} \quad (2.11)$$
$$Q_{\text{coll},i,j} = K_{i,j} N_{l} N_{i} \quad (2.12)$$

with $j = \text{NS, KS, AS, CS, KI, AI, CI}$. The collision kernels for liquid droplets, $K_{i,j}$, are taken for an average cloud droplet radius of 10 µm and for average aerosol mode radii. Variations of the actual diameters are neglected. The values are taken from Young (1974b) for growing droplets and are listed in Table 2.1. They represent lower estimates of the collision efficiency, which can be enhanced by thermophoretic effects. For example, the collision kernels calculated by Romakkaniemi et al. (2006) range between $10^{-13}$ and $10^{-9} \text{m}^2 \text{s}^{-1}$ for hydrometeors between 10 and 100 µm radius. For ice crystals, the shape dependency is neglected and values for the collision kernels for ice crystals, $K_{i,j}$, for an average crystal radius of 15 µm are estimated from Young (1974b). This simplified treatment is not expected to introduce a significant error, as impaction scavenging by ice crystals has been shown to be negligible for the in-cloud aerosol mass budget in a case study by Respondek et al. (1995).

The interstitial modes are also affected by droplet/ice nucleation, collisions, evaporation and sublimation. For the grid-mean number concentrations the prognostic equation is:

$$\frac{dN_{l,j}}{dt} = \frac{dN_{l,j}}{dt} \bigg|_{\text{transport}} + \frac{dN_{l,j}}{dt} \bigg|_{\text{aerosolmicrophysics}} + \frac{dN_{l,j}}{dt} \bigg|_{\text{sources}} + \frac{dN_{l,j}}{dt} \bigg|_{\text{sed,dep,bc-scav}} + f_j (\bar{r}_{\text{CD}}) Q_{\text{evap}} + f_j (\bar{r}_{\text{IC}}) Q_{\text{sub}}$$
$$- \frac{N_{\text{act},j}}{\sum_{k=1}^{4} N_{\text{act},k}} Q_{\text{nuc}} - \frac{N_{\text{act},j}}{\sum_{k=2}^{4} N_{\text{act},k}} Q_{\text{nuc,i}} - Q_{\text{coll},j} - Q_{\text{coll},i,j}$$

(2.13)

with $j = \text{NS, KS, AS, CS, KI, AI, CI}$. 

16
Table 2.1: Collision kernels $K_{i,j}$ for aerosol-droplet collisions and $K_{i,j}$ for aerosol-crystal collisions in m$^3$ s$^{-1}$ after Young (1974b), obtained for hydrometeors of 10 $\mu$m (droplets) and 15 $\mu$m (crystals) radius and the average aerosol size of the respective mode (see Figure 2.1).

<table>
<thead>
<tr>
<th>Mode</th>
<th>$K_{i,j}$</th>
<th>$K_{i,j}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nucleation soluble (mixed)</td>
<td>$2.5 \times 10^{-12}$</td>
<td>$5.0 \times 10^{-11}$</td>
</tr>
<tr>
<td>Aitken soluble (mixed)</td>
<td>$2.5 \times 10^{-12}$</td>
<td>$5.0 \times 10^{-11}$</td>
</tr>
<tr>
<td>Accumulation soluble (mixed)</td>
<td>$2.0 \times 10^{-14}$</td>
<td>$2.0 \times 10^{-12}$</td>
</tr>
<tr>
<td>Coarse soluble (mixed)</td>
<td>0.0</td>
<td>$2.0 \times 10^{-13}$</td>
</tr>
<tr>
<td>Aitken insoluble</td>
<td>$2.5 \times 10^{-12}$</td>
<td>$5.0 \times 10^{-11}$</td>
</tr>
<tr>
<td>Accumulation insoluble</td>
<td>$2.0 \times 10^{-14}$</td>
<td>$2.0 \times 10^{-12}$</td>
</tr>
<tr>
<td>Coarse insoluble</td>
<td>0.0</td>
<td>$2.0 \times 10^{-13}$</td>
</tr>
</tbody>
</table>

The second term on the right-hand side includes changes to the number concentration by aerosol microphysical processes, like inter- and intramodal coagulation and growth to larger modes through coating with sulfate. The third term takes into account particle emissions and nucleation from the gas phase (for mode NS), and the fourth term sedimentation, dry deposition and below-cloud scavenging. For evaporating cloud droplets and sublimating ice crystals, the dry number median radius of the residuals ($\bar{r}_{CD}$ and $\bar{r}_{IC}$, respectively) is calculated from grid-mean mass and number concentrations, assuming lognormal size distributions, as follows:

$$\bar{r}_{CD} = \sqrt[3]{\frac{\sum x m_{x,CD}}{4\pi \rho_{CD} \exp \left(\frac{2}{7} \ln^2 \sigma_{CD}\right) b N_i}}$$

$$\bar{r}_{IC} = \sqrt[3]{\frac{\sum x m_{x,IC}}{4\pi \rho_{IC} \exp \left(\frac{2}{7} \ln^2 \sigma_{IC}\right) b N_i}}$$

with $x = $ SO4, BC, OC, SS, DU.

The standard deviations of the cloud-borne modes, $\sigma_{CD}$ and $\sigma_{IC}$, are fixed to the same value as for the accumulation modes ($=1.59$), if the number median radius is smaller than 0.5 $\mu$m, or to the same values as for the coarse modes ($=2.0$) otherwise. The densities $\rho_{CD}$ and $\rho_{IC}$ are calculated as mass-weighted averages of the densities of the different components.

$$\rho_{CD} = \sum x \frac{m_{x,CD}}{\sum y m_{y,CD}} \rho_x$$

$$\rho_{IC} = \sum x \frac{m_{x,IC}}{\sum y m_{y,IC}} \rho_x$$

with $x, y = $ SO4, BC, OC, SS, DU.

The dry densities of the components are $\rho_{SO4} = 1841$ kg m$^{-3}$, $\rho_{BC} = \rho_{OC} = 2000$ kg m$^{-3}$, $\rho_{SS} = 2165$ kg m$^{-3}$ and $\rho_{DU} = 2650$ kg m$^{-3}$. The residuals are then attributed to the soluble mode with $\bar{r}_{CD}$ falling in its size range (given in Figure 2.1). Therefore the functions $f_j(\bar{r}_{CD})$ and $f_j(\bar{r}_{IC})$ in equation (2.13) are 1 if $\bar{r}_{CD}$ or $\bar{r}_{IC}$, respectively, lies in the size range of the soluble mode $j$, and 0 otherwise. Release to the nucleation mode is not allowed, because this mode contains only sulfate particles freshly nucleated from the gas phase.

Finally, the grid-mean masses of the different components in the interstitial modes change similar to equation (2.13):

$$\frac{dm_{x,j}}{dt} = \left| \frac{dm_{x,j}}{dt} \right|_{\text{transport}} + \left| \frac{dm_{x,j}}{dt} \right|_{\text{aerosol microphysics}} + \left| \frac{dm_{x,j}}{dt} \right|_{\text{sources}} + \left| \frac{dm_{x,j}}{dt} \right|_{\text{sed,dep,bc–scav}} + \frac{m_{x,CD}}{b N_i} f_j(\bar{r}_{CD}) Q_{\text{evap}} + \frac{m_{x,IC}}{b N_i} f_j(\bar{r}_{IC}) Q_{\text{sub}} - \frac{m_{x,j}}{N_j} \sum_{k=1}^{N_{\text{act},j}} \frac{N_{\text{act},k}}{N_{\text{act},j}} Q_{\text{nuc}}$$

17
2 Aerosol processing: SCM

\[ \frac{-m_{x,j}}{N_j} \sum_{k=2}^{4} \frac{N_{\text{acti},j}}{N_{\text{acti},k}} Q_{\text{nuci}} - \frac{m_{x,j}}{N_j} Q_{\text{coll},j} - \frac{m_{x,j}}{N_j} Q_{\text{coll},j} \]  

(2.18)

with \( x = \text{SO}_4, \text{BC}, \text{OC}, \text{SS}, \text{DU} \) and \( j = \text{NS}, \text{KS}, \text{AS}, \text{CS}, \text{KI}, \text{Al}, \text{Cl} \).

A few remarks about the underlying model assumptions: We assume that particles released from evaporating droplets are an internal mixture of all foreign material contained in the drop volume and that this newly formed particle does not break up. This is in agreement with wind-tunnel studies of droplets containing salts and insoluble particles by Mitra et al. (1992). For sublimating ice crystals, a similar process is presumed, and the processed material is released as one internally mixed particle. As the processed particles always contain some sulfate, which is produced in the aqueous phase, they are attributed to the soluble/mixed modes. Furthermore, we assume that upon droplet freezing, all foreign mass contained in the liquid is transferred into the ice crystal. As reviewed by Stuart and Jacobson (2003), the retention of soluble compounds during freezing is a function of their volatility. Volatile compounds can be repulsed from the solute when the solid ice phase is formed. However, as sulfate has a very low volatility, the assumption of complete retention seems to be valid.

### 2.2.3 SCM model setup

The ECHAM5-HAM single-column model is set up at the location of the Jungfraujoch, Switzerland (Figure 2.2). The single-column model requires initial vertical profiles of temperature, humidity, and wind. For this we used radiosonde data from either Payerne (Switzerland) or Milan (Italy). The values were prescribed for 3-5 timesteps and after that, if necessary, advective tendencies of temperature were applied in order to match observations at the Jungfraujoch. As the model orographic height is only 482m above sea level for the grid box including the Jungfraujoch coordinates, we compare the simulated fields on the appropriate pressure level (\( \approx 650 \text{ hPa} \)) to the observations.

We have selected three cases from the CLACE 3 (Cloud and Aerosol Characterization Experiment, see section 2.3) campaign (winter 2004) with different degrees of glaciation (as evident from the ice mass fraction). Criteria for the selection of these cases were that both total and interstitial aerosol size distributions were available before and after formation of the cloud and that during this time period no obvious change in airmass occurred. As the aim of this study is to analyze the evolution of the total and interstitial aerosol size distributions during cloud formation, changes in airmass disrupt the observation of this process and complicate the interpretation. However, as suitable cases were sparse and...
2.3 Observations of in-cloud and interstitial aerosol size distributions during CLACE 3

The Jungfraujoch is located on an exposed mountain col in the Swiss Alps at 3580m altitude (Figure 2.2). It is considered to be prevalently in the free troposphere, but can be influenced by convective intrusions from the planetary boundary layer, especially in spring and summer (Nyeki et al., 1998). The Jungfraujoch is often immersed in clouds (e.g. 33% of the time in March 2004, the month from which events for the present study were selected).

In addition to continuous aerosol and trace gas monitoring at the Global Atmosphere Watch (GAW) station and meteorological observations by the Swiss weather service MeteoSwiss, measurements during the CLACE campaigns provide a unique dataset of cloud-borne and interstitial aerosol size distributions. Henning et al. (2004) investigated size-dependent aerosol activation in mixed-phase clouds and found a dependency of the scavenged aerosol fraction on cloud temperature. Verheggen et al. (2007) extended this study, based on a larger database, and concluded that the decrease of the scavenged fraction with
Aerosol processing: SCM

Figure 2.3: Time series of LWC (liquid water content) and IWC (ice water content) observed at the Jungfraujoch during March 2004. The selected cases are marked with the letters A, B and C.

decreasing temperature below 0°C and with increasing ice mass fraction is mostly likely due to the evaporation of droplets in the presence of ice crystals (Wegener-Bergeron-Findeisen process). Cozic et al. (2007) focused on the scavenging of black carbon, which was found to be scavenged into the liquid cloud phase to the same extent as the bulk aerosol. This suggests that the majority of the black carbon containing particles at the Jungfraujoch are covered with soluble material through aging processes.

Measurements during CLACE 3 are described in detail by Verheggen et al. (2007); a short summary is given below. The particle size distributions were obtained behind two different inlets, a total inlet and an interstitial inlet. The total inlet samples both interstitial particles and residuals from cloud droplets and ice crystals, which are evaporated by heating the inlet to 25°C. The interstitial inlet removes cloud particles larger than 2 µm. Alternating behind the two inlets, a Scanning Mobility Particle Sizer (SMPS) measured the particle size distributions between radii of 8.5 to 450 nm every 6 minutes under dry conditions. The difference in concentration obtained behind the total and the interstitial inlet is considered representative of the cloud-borne particle concentration.

Cloud liquid water content was measured at a 1-minute time resolution with a Particle Volume Monitor (PVM) (K. Bower, M. Flynn and P. Connolly, personal communication). The values are empirically corrected for the presence of ice and averaged over 12 minutes. Ice water content is derived from the size spectrum and 2-dimensional images of hydrometeors measured by a Cloud Particle Imager (CPI) using the procedure described by Connolly et al. (2007). The error in ice water content is estimated to be up to 50%. The error in liquid water content depends on the amount of coexisting ice, which increased the uncertainty of the measurement. For the cases studied here, the possible error ranges between 5 and 40%, with lower errors for a small ice water content.

Precipitation was not measured in situ at the Jungfraujoch. We show precipitation estimates extracted from a gridded dataset based on radar measurements (Wüest et al., subm.).

2.4 SCM results for CLACE mixed-phase clouds

In the following, simulations for three cases of mixed-phase clouds are described. These cases differ mainly in the ice mass fraction and the cloud evolution. Figure 2.3 shows time series of the measured LWC (liquid water content) and IWC (ice water content) for the whole month of March 2004. Periods A, B, and C are indicated.

2.4.1 Case A: liquid cloud (March 30)

For the first case studied here, we focus on the mainly liquid part of a cloud event on March 30, 2004, between 16:24 UTC and 17:12 UTC, with an ice mass fraction of 15 – 36%. This event is chosen to
demonstrate the model’s behavior for a relatively simple case, for which continuous data are available for comparison.

Total and interstitial observed size distributions, LWC and IWC are shown in Figures 2.4(a) and (b). Figure 2.6 gives the evolution of the total and interstitial concentrations integrated over all sizes.

Before 15:00 UTC, low LWC but high IWC was observed. Then, after a short cloud-free period around 15:30 UTC, the LWC reaches 0.08 g m\(^{-3}\) during the liquid cloud period (16:24-17:12 UTC). As the temperatures range between \(-7\) to \(-10^\circ\)C, the liquid water can not result from melting of ice, but only from droplet nucleation from the vapor phase. The ice water content has values \(< 0.04 \text{ g m}^{-3}\) after 16:24 UTC. Moreover, the lower 15%-percentile of the IWC (not shown) is \(0 - 0.01 \text{ g m}^{-3}\), which points to a mainly liquid cloud with a few ice crystals. The difference between observed total and interstitial aerosol concentration is rather small (\(< 100 \text{ cm}^{-3}\)) before 16:24 UTC, indicating low hydrometeor number concentrations (due to the low LWC) and a low fraction of particles incorporated in the ice crystals. Upon the formation of the liquid cloud around 16:24 UTC, the interstitial aerosol number decreases from 800 cm\(^{-3}\) to 200 cm\(^{-3}\). The difference between observed total and interstitial aerosol number (which is an estimate of the hydrometeor concentration) increases to 280 cm\(^{-3}\). The total aerosol concentration decreases only slightly during cloud formation. Thus, it seems reasonable to assume cloud formation on the same population of aerosol particles as measured before cloud (i.e. between 14:00 UTC and 16:00 UTC). The decrease in the concentration of small particles can be due to collision scavenging. Larger particles activate to cloud droplets and are therefore removed from the interstitial phase, but remain visible in the total aerosol concentration. As no precipitation was observed by radar, it is not clear whether the partial cloud dissociation around 17:30 UTC is due to evaporation, slight local precipitation or advection of a less dense part of the cloud.

The model was initialized with a radio sounding from Milan, 18 UTC of March 30 (Figure 2.5). Two moist layers below a strong inversion were advected northwards. A small cooling rate of \(-0.25 \text{ Kh}^{-1}\), as it has been observed at the Jungfraujoch, was imposed at all levels. The aerosol concentrations were initialized with a bimodal fit to the mean total aerosol size distribution between 14:00 UTC and 16:00 UTC. In ECHAM5-HAM, liquid water starts to condense around 15:30 UTC, and the grid-mean LWC slowly increases to 0.02 g m\(^{-3}\) at 16:30 UTC. This is only 25% of the observed LWC. The model does not form any ice, as heterogeneous freezing is not efficient at these warm subzero temperatures. Precipitation does not form either. At 17:15 UTC, cloud dissipation by evaporation has been imposed.

Figure 2.6 shows that the simulated interstitial aerosol concentration steadily decreases as cloud droplets nucleate, while the total aerosol concentration remains relatively constant. This is similar to observations, although the process is slower in the model. Additionally, the observations show a decrease in the total aerosol concentration during the liquid cloud phase, for which the cause is not clear. The difference between the simulated total and the interstitial aerosol number concentration gives the simulated cloud droplet number, which reaches the maximum observed value at 16:45 UTC. The total aerosol size distribution (Figure 2.4(c)) changes its shape because aerosol particles from the Aitken mode, with \(\sigma = 2.44\), and from the accumulation mode, with \(\sigma = 1.66\) (standard deviations obtained from the fit), are transferred into one in-cloud mode with \(\sigma = 1.66\). The radius of the in-droplet mode is the weighted average of the Aitken and accumulation mode contributions. The total aerosol remains unchanged upon cloud evaporation, when the in-droplet aerosol is transferred back into the interstitial accumulation mode. In-cloud sulfate production is negligible for this case, as the prescribed SO\(_2\) concentration is very low (0.11 ppbm, daily mean value for March 30, 2004).

The total aerosol size distribution at the end of the simulation differs from the initial state. This is due to collision-coalescence processes and to the representation of in-droplet particles by only one mode, into which the properties of all activated particles are averaged. It is difficult to estimate the error introduced by this assumption, as the observed total aerosol size is not recovered either. This can be the case for a number of reasons: droplet collision-coalescence reduces the observed number (but not mass) of cloud-borne particles, in-cloud sulfate production adds to the cloud-borne mass, and precipitation formation reduces both in-droplet aerosol number and mass.

For comparison, Figures 2.4(e) and (f) show the evolution of the size distribution in the standard ECHAM5-HAM without explicit treatment of cloud-borne particles. As no precipitation forms, the total aerosol number concentration remains constant. A prescribed fraction of 25% (mass and number) for the Aitken mode and of 85% for the accumulation mode is considered cloud-borne in the cloudy fraction of the gridbox, which increases here from 0 to 0.6. The standard model can not reproduce the changes in the total aerosol size distribution by cloud processing, and for this case the interstitial aerosol concentration is overestimated.
Figure 2.4: Observed ((a) and (b)) and simulated ((c) and (d)) total and interstitial aerosol size distributions for case A. Red lines are precipitation (left plots, right axis; zero in this case) and liquid water content (LWC, continuous line, right axis, right plots) and ice water content (IWC, dashed line, right axis, right plots). Plots (e) and (f) show the total and interstitial aerosol size distributions diagnosed from the standard version of ECHAM5-HAM (without explicit treatment of cloud-borne particles).
2.4 SCM results for CLACE mixed-phase clouds

Figure 2.5: Initial sounding for case A and in-situ observations of the temperature $T$ and the dewpoint $T_d$ at the Jungfraujoch.

Figure 2.6: Observed and simulated total and interstitial aerosol number concentrations for case A.
2.4.2 Case B: glaciating cloud (March 10)

In this case, a cloud is observed which first consists mainly of liquid water and later predominantly of ice. The observed LWC, IWC, precipitation and aerosol size distributions are plotted in Figure 2.7. Obviously, the appearance of the cloud at the Jungfraujoch around 5:00 UTC was associated with a change of airmass, as the total aerosol concentration also increased significantly around that time. Between 5:00 UTC and 6:20 UTC, the cloud is nearly completely liquid with an ice mass fraction of 0 − 6%. Then within approximately 30 minutes the ice mass fraction increases to \( \approx 90\% \). This can be associated with cloud glaciation and/or another change of air mass. As the total aerosol distribution does not show any abrupt changes during this time, it is likely that a main contribution to the increase in ice is due to in-situ freezing. Radar observations show significant precipitation (up to 0.7 mm h\(^{-1}\)) in the Jungfraujoch area, suggesting that removal of aerosols by washout and rainout occurred. The contrary observation of a steady, continuous increase of total aerosol during the whole cloudy period despite the precipitation scavenging can only be explained with the advection of aerosol-rich air.

From 5:24 UTC to 6:24 UTC (while the cloud consists of supercooled liquid water), the observed total and interstitial aerosol size distributions differ clearly. As can be seen in Figures 2.7 and 2.8 (a), the total aerosol and interstitial size distributions are very similar for small particles, but the total aerosol concentration is larger than the interstitial concentration for \( r > 40 \text{nm} \). This reflects the preferential activation of larger aerosol particles. The scavenged fraction is here defined as

\[
F_N(r > 50 \text{nm}) = \frac{N_{\text{tot}}(r > 50 \text{nm}) - N_{\text{int}}(r > 50 \text{nm})}{N_{\text{tot}}(r > 50 \text{nm})}
\]  

(2.19)

following Verheggen et al. (2007) (who use the term ‘activated fraction’), with \( N_{\text{tot}}(r > 50 \text{nm}) \) the total aerosol number concentration integrated over radii larger than 50nm and \( N_{\text{int}} \) the interstitial aerosol number concentration integrated over radii larger than 50nm. Note that \( F_N \) includes both nucleation scavenging (activation of a cloud droplet on an aerosol particle) and collision scavenging (incorporation of an aerosol particle into a hydrometeor through impaction). \( F_N \) is shown in Figure 2.10. The observed scavenged fraction is approximately 0.6 from 5:24 UTC to 6:24 UTC. Upon the increase in ice mass fraction, the partitioning of the aerosol particles between the interstitial and cloud-borne phases changes. The scavenged fraction decreases to values below 0.2. In Figure 2.7, no clear size dependence of the difference between total and interstitial concentrations can be seen in the glaciated cloud (after 7:45
2.4 SCM results for CLACE mixed-phase clouds

Figure 2.8: Size distributions in case B, liquid part, (a) from observations and from simulations (b) with and (c) without collision scavenging of aerosol particles by cloud droplets.

Figure 2.9: Initial sounding for case B and in-situ observations of the temperature $T$ and the dewpoint $T_d$ (identical from 970 to 610 hPa) at the Jungfraujoch.
2 Aerosol processing: SCM

Figure 2.10: Evolution of the scavenged fraction $F_N$ for case B. ECHAM5-HAM with/without aerosol-droplet collisions refers to the new version with explicit treatment of in-cloud modes, while the scavenged fraction from prescribed scavenging parameters is calculated with the standard version of the model.

UTC). This behavior can be explained with the release of cloud-borne particles from evaporating droplets back into the interstitial phase during the Wegener-Bergeron-Findeisen process (Verheggen et al., 2007).

The model has been initialized with two lognormal aerosol modes, fitted to the total aerosol distribution in the liquid part of the cloud (see Figure 2.8), as the total aerosol before cloud was clearly not representative for the aerosol on which the cloud has formed. The temperature, humidity and wind velocity profiles were taken from a radio sounding from Milan from 6 UTC of March 11 (Figure 2.9), as the soundings from March 10 were too dry to simulate cloud formation at the Jungfraujoch gridpoint. The sounding from March 11 is moister and 2–3°C warmer than the temperature measured at the Jungfraujoch, which varied between $-22$ and $-18^\circ$C during the considered cloud event. Similar to the observed event, the model also produces a cloud which is first liquid and then glaciates. The LWC is in the same order of magnitude as the measurements ($0.19$ g m$^{-3}$ compared to $0.3$ g m$^{-3}$), but the IWC reached in the model is only $0.1$ g m$^{-3}$, while the observed IWC reaches $0.6–0.8$ g m$^{-3}$. This can be related to the depletion of humidity in the model by precipitation and the missing advection of humid air.

During the liquid cloud period, a large fraction of the available aerosol in the model is either activated or scavenged via collisions (Figure 2.7). To separate these two processes, a simulation without aerosol-droplet collisions has been run. The difference in the resulting size distributions as snapshots at 5:30 UTC is shown in Figure 2.8. It can be seen that aerosol activation alone mainly removes the large particles from the interstitial phase, while the small particles are scavenged by the collisions.

The collision coefficients chosen here represent rather a lower limit of estimates in the literature. Thus it is surprising that no depletion of small particles is found in the measurements. Solving equation 2.11, disregarding all other processes, gives $N_j(t + dt) = N_j(t) \exp \left( -K_{l,j} N_l dt \right)$. For the Aitken mode in this example, with $N_l = 590$ cm$^{-3}$ and $K_{l,j}$ from Table 2.1, this yields a reduction of the interstitial aerosol concentration by 8.5% per minute. Such a decrease is not observed. As the measurements are averages over six minutes, a heterogeneous cloud structure, with patches of low cloud droplet and high aerosol concentration in the small size range, might explain this bias. However, total number concentration measurements at a higher time resolution (not shown) do not show any rapid fluctuations. It is also possible that in this case an orographic cloud was observed, which was freshly formed close to the Jungfraujoch and that the observed air parcels had only a very short in-cloud history. Furthermore, the total aerosol distribution during the cloud is not identical to the unknown before-cloud size distribution, which might have contained more smaller particles. In addition, it is possible, but rather unlikely, that the small particles are refilled from an unknown source. Finally, the collision coefficients derived from theoretical considerations for a droplet size of 10 µm radius, specified temperature and relative humidity, may not be applicable to this case with an unknown droplet size distribution and unknown turbulent and phoretic contributions to the collision efficiency. The mean droplet size between 5:24 UTC to 6:24 UTC, derived from the liquid water content and the cloud drop number concentration, lies between 4.3 and 5 µm. For smaller droplets the collision efficiency is smaller, which is not taken into account in the model.
2.4 SCM results for CLACE mixed-phase clouds

Figure 2.11: Change in number concentrations of total and interstitial aerosol particles for case B. The large values before 5:30 UTC are probably associated with a change in airmass, while the peak in interstitial aerosol tendency around 7:00 UTC is attributed to the release of interstitial aerosol particles during glaciation. The dotted line is the average over the positive values of the total aerosol number concentration change after 6:00 UTC and gives a lower estimate for the advection term.

The simulated scavenged fraction in the model rapidly goes up to 0.8 (Figure 2.10). As the scavenged fraction $F_N$ refers to particles with radii larger than 50 nm, which are less affected by collision scavenging, the difference between the simulations with and without collision scavenging is relatively small. The Wegener-Bergeron-Findeisen process is implemented in ECHAM5-HAM in a way that all liquid water is evaporated instantaneously as soon as a threshold of IWC is passed. Therefore the scavenged fraction drops to very low values when the cloud glaciates. The ice crystal number concentration after glaciation is initially 16 cm$^{-3}$ and later even drops to < 100 cm$^{-3}$ (not shown). The difference between measured total and interstitial aerosol, which provides an estimate of the hydrometeor concentration, still amounts to 50–250 cm$^{-3}$ after glaciation. As the observed LWC is small but positive, the hydrometeor number concentration probably contains many liquid droplets. Again, this might be related to cloud heterogeneity and the presence of liquid patches and could explain the underestimation of the scavenged fraction in the ice cloud period.

For comparison, the scavenged fraction diagnosed for the standard ECHAM5-HAM model (without explicit treatment of cloud-borne particles) is also included in Figure 2.10. The prescribed scavenging parameters vary between 0.4 and 0.85 for Aitken and accumulation mode particles in liquid and mixed-phase clouds (Stier et al., 2005). The average scavenged fraction is weighted with the relative mass contribution of these two modes and slowly decreases from 0.8 to 0.65. This treatment does not capture any gradual increase of the scavenged fraction during cloud formation nor the strong decrease upon glaciation.

In an attempt to analyze the importance of aerosol advection for this case, Figure 2.11 shows the changes in number concentrations of the total ($dN_{\text{tot}}/dt$) and the interstitial ($dN_{\text{int}}/dt$) aerosol. For $dN_{\text{tot}}/dt$, this change represents the sum of advection and losses by impaction scavenging and precipitation, and for $dN_{\text{int}}/dt$, the sum of advection, incorporation into and release from hydrometeors and loss by impaction scavenging. The large values of both $dN_{\text{tot}}/dt$ and $dN_{\text{int}}/dt$ before 5:30 UTC are probably associated with a change in airmass. From 6:00 UTC to 9:15 UTC, the change in number concentration is on average positive, which signifies that advection of aerosol particles overcompensates the (unknown) loss terms. The mean of the positive values of $dN_{\text{tot}}/dt$ is 0.53 cm$^{-3}$ s$^{-1}$, which can be interpreted as a lower limit for the contribution of advection to the change in $N_{\text{tot}}$. In $dN_{\text{int}}/dt$, the peak around 7:00 UTC during cloud glaciation stands out against the relatively low advection from 6:00 UTC to 9:15 UTC, suggesting that a process-based interpretation of the evolution of the aerosol concentration is reasonable. As a sensitivity study, the average advection term has been added continuously to the simulated interstitial aerosol (Figure 2.12)). This has only a small influence on the aerosol depletion during the liquid cloud period (5:15 to 6:00 UTC), which is stronger in the model than in the observations. Later on, the interstitial aerosol is partly replenished.

The gas-phase SO$_2$-concentration (0.75 ppbm), again prescribed from GAW measurements, was markedly higher on March 10, 2004 than for case A. In case B in-cloud sulfate production becomes significant and
Figure 2.12: Observed and simulated total and interstitial aerosol number concentrations for case B. For the model simulation plotted in blue, the advection term estimated from Figure 2.11 has been added continuously after model initialization.

contributes 12% to the peak in-droplet sulfate mass. This contribution is lower than the global average contribution of 50-60% given by Barth et al. (2000), which is not surprising as the air at the Jungfraujoch is very clean.

2.4.3 Case C: coexistence of liquid and ice (March 20)

In the night from March 19 to March 20, starting at 23:00 UTC, a mixed-phase cloud was observed which showed different properties than the cases described above. The temperature ranged between -8 and -4°C. Before 23:00 UTC, although some LWC and IWC has been observed, the lower 15-percentiles, based on 1-minute-averages, are 0.01 g m$^{-3}$, which hints at a thin, heterogeneous cloud with holes. Between 23:00 UTC and 0:00 UTC, the cloud consists mainly of ice, and after 1:00 UTC, the ice mass fraction varies between 30-80%. Precipitation of up to 0.1 mm h$^{-1}$ was observed. The aerosol size distributions (Figures 2.13(a) and (b)) show high and similar total and interstitial aerosol concentrations before 0:00 UTC. The strong peak around 23:00 UTC, especially at very small and large radii, might be related to local pollution e.g. by night-time construction work and has been omitted from further analysis. The interstitial aerosol concentration rapidly decreases as soon as the LWC increases. At the same time the total aerosol also decreases, but much slower. The decrease in total aerosol is either linked to coagulation, precipitation scavenging or to a change in airmass.

This case is rather unusual, as ice mass fractions between 0.2 and 0.8 occurred at a lower frequency during the CLACE campaigns than high or low values (close to pure ice/pure liquid conditions) (Verheggen et al., 2007). This has also been reported previously for a dataset of aircraft measurements by Korolev et al. (2003). Supercooled liquid water and ice can not coexist in a stable equilibrium, as the saturation vapor pressure over ice is lower than over water. This would lead to evaporation of the droplets. Non-equilibrium conditions might however be possible if an updraft is strong enough to provide enough supersaturation over water to balance the evaporation (Korolev, 2007). Furthermore, pockets of pure liquid and pure ice on scales in the orders of kilometers (Korolev et al., 2003) and down to tens of meters (Chylek and Borel, 2004) have been observed in clouds which on average appear to have an intermediate ice mass fraction. As the average winds during this case are about 9 m s$^{-1}$, during a 12-minute sampling period a region of, on average, 6.5 km is probed. Hydrometeor shapes were measured by a Cloud Particle Imager (CPI) with a time resolution of 1 second. For case C, concentrations of spherical particles (which are assumed to be liquid) and of ice crystals in various shapes anticorrelate, which hints at possible cloud heterogeneity (K. Bower, M. Flynn and P. Connolly, personal communication). Therefore this case is difficult to understand and also to model appropriately.

The results of a simulation with the standard cloud microphysical configuration are shown in Figure 2.13(c) and (d). ECHAM5-HAM was initialized with a sounding from Payerne, 0:00 UTC on March 20 (Figure 2.14), and with a bimodal fit to the total aerosol concentration between 17:00-22:30 UTC. The model behaves similarly to case B: First a liquid cloud is formed, with LWC increasing up to 0.12 g m$^{-3}$. During this liquid period the interstitial aerosol is partly scavenged by activation and collisions. Then, at
2.4 SCM results for CLACE mixed-phase clouds

Figure 2.13: Observed and simulated total and interstitial aerosol size distributions for case C. Red lines are precipitation (left plots, right axis) and liquid water content (LWC, continuous line, right axis, right plots) and ice water content (IWC, dashed line, right axis, right plots). (c) and (d): with cloud glaciation via the Wegener-Bergeron-Findeisen process, (e) and (f): with forced non-equilibrium and coexistence of liquid and ice.
22:00 UTC, glaciation sets in and the remaining liquid water is evaporated immediately. This also leads to release of cloud-borne aerosol back into the interstitial phase. During the following hours, simulated hydrometeor number concentrations are very low. Total and interstitial aerosol concentrations are both slowly decreasing due to precipitation formation. However, the precipitation does not reach the ground (and is therefore 0.0 on Figure 2.13(c)) because it evaporates in drier below-cloud layers.

As a sensitivity study, it has been tested whether a different parameterization of the Wegener-Bergeron-Findeisen process would lead to a better representation of the coexisting phases. For this purpose, a higher humidity has to be imposed (0.001% supersaturation over water during the whole period). This induced liquid water condensation at all timesteps. Additionally, in contrast to the standard model, we allowed ice deposition after consumption of the vapor by condensation down to 100% relative humidity with respect to water. In cases where simultaneous deposition and condensation occurred, the Wegener-Bergeron-Findeisen process was switched off. Simultaneous deposition and condensation can only occur in a non-equilibrium situation, when e. g. an updraft is strong enough to provide supersaturation over water before the vapor is consumed by deposition to the ice phase. The results of this simulation are shown in Figure 2.13(e) and (f). We achieved coexistence of ice (0.03 g m\(^{-3}\)) and liquid water (0.1 g m\(^{-3}\)), accompanied by a faster and more complete depletion of the interstitial aerosol. Especially the smaller particles are removed faster, because the hydrometeor concentrations are higher. This depletion is closer to the observations than the release of interstitial aerosol associated with the cloud glaciation described above. However, we can not be sure whether the observed coexistence is caused by a strong updraft providing supersaturation over water or a result of cloud heterogeneity.

2.5 Summary and conclusions

A novel description of cloud-borne aerosol particles and their processing in the framework of the global aerosol-climate model ECHAM5-HAM is presented. Other than previous studies with global aerosol models, particles included in cloud droplets and particles in ice crystals are treated separately, which allows accounting for the influence of mixed-phase cloud processes on aerosol concentration, composition, size distribution and mixing state. Furthermore, this feeds back on subsequent cloud formation.

In addition to the seven aerosol modes in HAM, two new modes representing in-droplet and in-crystal
particles have been introduced. Their numbers are coupled to the number of cloud droplet and ice crystal concentrations simulated by the ECHAM5 double-moment cloud microphysics scheme.

Observations during CLACE at the high-altitude station Jungfraujoch allow validation of the transfer and loss mechanisms of interstitial and cloud-borne aerosol. Three different mixed-phase cases have been studied with the single column model version of ECHAM5-HAM. During periods dominated by liquid water (case A and first part of case B), the model simulates incorporation of a large fraction of the interstitial aerosol into the cloud droplets. Larger particles are activated, while smaller particles are scavenged via collisions. The scavenged fraction in the liquid period increases more slowly than observed in case A, but is higher than observed in case B, when especially the collision scavenging of small particles seems to be overestimated. This is probably related to uncertainties in the simulated cloud properties (cloud droplet number concentration, updraft velocities), or to too high values of the collision efficiencies for this case and to subgridscale variability.

Rapid cloud glaciation was observed in case B. The model simulated a similar cloud evolution with complete cloud glaciation via the Wegener-Bergeron-Findeisen process. During this transition, cloud droplets evaporate and the particles contained or dissolved in them are released back into the interstitial phase. This is seen in both the measurements and the model results. However, while the simulated cloud consists only of ice and has a very low hydrometeor concentration, in reality some liquid water remains and the scavenged fraction does not decrease as strongly as in the model.

In case C, during the 12-minute sampling period, a mixture of ice and supercooled liquid water is observed. This can not be captured by the model. In the standard model version a liquid cloud which freezes completely (similar to period B) is simulated. Therefore the scavenged fraction is too low. If a higher relative humidity is imposed and the cloud microphysical parameterizations are modified in a way that allow coexistence of supercooled water and ice, the scavenged fraction remains very high. It can not be distinguished from the measurements whether the seemingly coexisting liquid and water does really occur simultaneously, because a strong updraft supplies enough humidity to balance the diffusion of water vapor from droplets to ice crystals, or whether the cloud consists of a heterogeneous mixture of liquid and ice-phase clusters. Neither scenario can be captured in a model which runs in a resolution of tens to hundreds of kilometers.

We conclude that the model qualitatively represents the important transfer processes between interstitial and cloud-borne aerosol in mixed-phase clouds (droplet activation, collision scavenging, retention during freezing, release during evaporation) in a reasonable way. Quantitative comparisons between the model and the observational data were hampered by uncertainties in the environmental conditions and lack of information about airmass changes and particle/cloud hydrometeor advection. The model simulates high transfer rates into hydrometeors in liquid cloud phases by activation and collision scavenging, and a nearly complete release during cloud glaciation. In-cloud sulfate production is relatively small in the cases studied here, because the gas-phase SO$_2$-concentrations at the Jungfraujoch are low. A higher contribution to the cloud-borne mass is expected for more polluted areas. In the future, this extended version of ECHAM5-HAM, which includes an explicit representation of aerosol processing, will be used for global simulations, allowing a global assessment of the influence on cloud processing on aerosol budgets and size distributions.

Acknowledgments

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3 Global simulations of aerosol processing in clouds

3.1 Introduction

Pruppacher and Jaenicke (1995) estimated that globally averaged an atmospheric aerosol particle, sampled at a distance from a specific source, has been cycled three times through a cloud. Uptake into cloud droplets, collision-coalescence, chemical processing inside hydrometeors and release back into the atmosphere has important implications for the physical and chemical properties of the aerosol.

In detail, a cycle of an aerosol particle through a liquid cloud can involve the following processes: Preferably the bigger and hygroscopic aerosol particles act as the cloud condensation nuclei on which cloud droplets form. Cloud droplets can collect more aerosol particles and other cloud droplets by collisions. The soluble part of the aerosol particles (e. g. sulfate, salts) dissolves in the water. Additionally, atmospheric gases can also transfer into droplets and undergo chemical reactions in the aqueous phase. E. g., the major part of atmospheric sulfate mass is formed from reactions inside cloud droplets (Barth et al., 2000). If precipitation is formed, all material collected in the precipitating droplets is removed (scavenged) from the atmosphere. However, a large fraction of clouds does not form precipitation, but evaporates (Lin and Rossow, 1996). In this case the dissolved material concentrates in the liquid phase again or crystallizes, and together with the possible insoluble material contained inside the droplet forms one new, internally mixed aerosol particle (Pruppacher and Klett, 1997). These reemitted particles are larger than prior to cloud processing.

Effects of cloud processing on the aerosol size distribution have been observed by Hoppel et al. (1986) and Hoppel et al. (1990). Marine boundary layer aerosol size distributions were found to exhibit a distinct bimodal shape. The second (larger) peak is attributed to activated particles which have grown through cloud processing, while freshly nucleated, not activated particles constitute the smaller mode. Bower et al. (1997) observed significant modifications of the aerosol size distribution and hygroscopic properties by the passage through a hill cap cloud. Addition of sulfate mass often increased the number of cloud condensation nuclei available for subsequent cloud formation.

In this chapter we apply the extended aerosol-climate model, as introduced in chapter 2, to global simulations of aerosol processing in clouds. The model includes prognostic equations for in-droplet and in-crystal aerosol mass.

3.2 Model description

ECHAM5-HAM (Stier et al., 2005) is a global aerosol-climate model with a prognostic treatment of cloud droplets and ice crystals (Lohmann et al., 2007). Both the cloud droplet activation and the ice crystal formation through homogeneous and heterogeneous freezing depend on the simulated aerosol number concentration, size distribution and composition.

The aerosol module HAM represents the atmospheric aerosol in seven internally and externally mixed modes, consisting of the five components sulfate, black carbon, organic carbon, sea salt and mineral dust. Hoosie et al. (2008b) have extended HAM by two additional modes which include in-droplet and in-crystal particles, respectively. A detailed model description of this aerosol processing can be found in chapter 2. The aerosol processing is now applied globally. Contrary to the single column model studies in chapter 2, aerosol, cloud droplet and ice crystal vertical and horizontal transport, vertical diffusion and aerosol and ice crystal sedimentation are now included. Advection and diffusion of in-droplet and in-crystal aerosol mass and the corresponding droplet and crystal numbers can be inconsistent if different gradients exist, and can lead to unrealistic sizes of the cloudborne particles. Cloudborne particles with a dry radius smaller than 5 nm or larger than 50 µm are removed. It was carefully examined that the global aerosol mass budgets are closed and no significant aerosol mass losses occurred.

Below-cloud evaporation is treated in the following simplified way: The mass fluxes of precipitation and the incorporated aerosol of each species are computed, and in each below-cloud level the fraction
of evaporating precipitation is calculated as a function of the relative humidity. The same fraction is applied to the release of aerosols from the evaporating hydrometeors, i.e. here an inhomogeneous mixing assumption is applied, in contrary to in-cloud evaporation. The aerosol mass from the evaporating precipitation particles is attributed to the soluble/mixed coarse mode. As no rain droplet number is computed in ECHAM5-HAM, the number of aerosol particles resulting from the evaporation of precipitation is estimated from the evaporating precipitation mass with the assumption of a radius of 50 \( \mu m \) per rain droplet. In the standard model version without explicit aerosol processing, particles are released back into the mode from which they were previously scavenged, i.e. their composition and size distribution remains unchanged.

Table 3.1 lists the simulations which are compared and discussed in this chapter. Simulation CTL is similar to the reference simulation described by Lohmann et al. (2007), with minor updates and corrections (Lohmann, 2007). For autoconversion, the process of transformation of cloud droplets to rain droplets, a different parameterization is used in this study. Lohmann et al. (2007) used Khairoutdinov and Kogan’s (2000) scheme, which has the following form:

\[
\frac{dN_l}{dt}_{\text{aut}} \equiv Q_{\text{aut}} = -\gamma 0.0525 q_{\text{air}}^{-1.47} q_l^{4.47} N_l^{-1.79} \tag{3.1}
\]

\[
\frac{dN_l}{dt}_{\text{aut}} \equiv Q_{\text{aut}} = \frac{N_l dq_l}{q_l dt} = -\gamma 0.0525 q_{\text{air}}^{-1.47} q_l^{1.47} N_l^{-0.79} \tag{3.2}
\]

Here the liquid water content \( q_l \) has to be given in g m\(^{-3} \), the cloud droplet number concentration \( N_l \) in cm\(^{-3} \), and the air density \( \rho_{\text{air}} \) in kg m\(^{-3} \). \( \gamma \) is a model tuning parameter, as described below. This parameterization has the disadvantage that it does not provide a term for selfcollection, i.e. cloud droplet growth which does not lead to precipitation yet. As previous studies (Flossmann et al., 1985; Ivanova and Leighton, 2008) have shown that droplet collision-coalescence leads to important redistributions of the in-droplet aerosol, we revert to the following parameterization by Beheng (1994) in order to include this process.

\[
\frac{dN_l}{dt}_{\text{aut+self}} \equiv Q_{\text{aut} + Q_{\text{self}}} = -\gamma 75.7 q_l^{4.7} N_l^{-3.3} \tag{3.3}
\]

\[
\frac{dN_l}{dt}_{\text{aut+self}} \equiv Q_{\text{aut} + Q_{\text{self}}} = 7.7\frac{dq_l}{dt} + Q_{\text{self}} = -\gamma 582.9 q_l^{4.7} N_l^{-3.3} - 0.013 q_l^2 \tag{3.4}
\]

The coefficients are valid for \( q_l \) in g m\(^{-3} \) \( N_l \) in cm\(^{-3} \). Beheng’s (1994) scheme was previously used in the ECHAM4 GCM (Lohmann and Roeckner, 1996; Lohmann et al., 1999). The parameter \( \gamma \) is used to scale the conversion of cloud liquid water to rain in such a way that a balanced radiation budget at the top of the atmosphere is achieved. A higher value of \( \gamma \) increases precipitation formation and lowers the global mean droplet number concentration and liquid water path, therefore decreasing the reflected shortwave radiation. This tuning is necessary and justified because subgrid-scale variations in the cloud droplet number concentration can not be resolved in global models, but can have a strong impact on the nonlinear process of rain formation. For the sensitivity simulations in Table 3.1, \( \gamma \) ranges between 15 and 400.
3.3 Comparison to the standard model

3.3.1 Global and zonal mean cloud and aerosol parameters

Table 3.2 gives an overview over global annual mean values of cloud-related variables. The global mean liquid water path is 67 g m\(^{-2}\) in simulation CTL, and 38 g m\(^{-2}\) in simulation AP. As can be seen in Figure 3.1, CTL lies in the upper range of values retrieved from satellite data (Greenwald et al., 1993; Wentz, 1997) except for the tropics, while AP follows more closely the retrieval by Weng and Grody (1994). The ice water path is similar in all simulations \((\approx 22 \text{ g m}^{-2})\), which is somewhat lower than an estimate from ISCCP data by Storelvmo et al. (2008) of 29 g m\(^{-2}\). The shortwave cloud forcing (SCF) is \(-55.8 \text{ W m}^{-2}\) in simulation CTL and \(-50.1 \text{ W m}^{-2}\) in simulation AP, compared to ERBE retrievals (Kiehl and Trenberth, 1997) of \(-50 \text{ W m}^{-2}\). Figure 3.1 (b) illustrates that the zonal distribution of the SCF is well captured in both simulations. The longwave cloud forcing, which depends mainly on ice clouds, is similar in all simulations and close to the observed value of approximately 30 W m\(^{-2}\). An equilibrated radiation budget \((F_{\text{net}} \approx 0)\) is achieved in all simulations (except AP-untuned), which is included in Table 3.2 for illustration of the effect of model tuning.

Figure 3.1 (c) reveals that the aerosol optical depth (AOD) is significantly higher in simulation AP (global mean 0.35) than in simulation CTL (0.19) and as obtained from observations (0.15 - 0.19). CTL agrees generally well with the observations, but overestimates the AOD at southern latitudes. Overestimation of aerosol optical depth can be due to several reasons: a too high aerosol mass burden, mis-predicted aerosol size distributions or an incorrect parameterization of aerosol optical properties. The aerosol burden and size distribution are significantly different with the new treatment of aerosol processing, as discussed below. Resulting from the differences in the atmospheric aerosol, the droplet number burden \(N_l\) is also higher in simulation AP than in simulation CTL (with a grid-mean value of \(2.7 \times 10^{10} \text{ m}^{-2}\)), although tuning of the autoconversion rate has reduced it from 4.6 to \(2.8 \times 10^{10} \text{ m}^{-2}\).
Figure 3.1: Annual zonal mean (a) liquid water path over ocean, (b) shortwave cloud forcing, (c) aerosol optical thickness, and (d) cloud droplet number burden in simulations AP and CTL. Black lines are different satellite observations: (a) SSM/I retrievals by Wentz (1997) (continuous line), Greenwald et al. (1993) (dashed line) and Weng and Grody (1994) (dash-dotted line), (b) ERBE measurements (Kiehl and Trenberth, 1997), (c) a combined MODIS/MISR retrieval (S. Kinne, personal communication), and (d) a retrieval based on ISCCP data (Han et al., 1998).

In a previous publication (Lohmann et al., 2007), this value has been compared to a retrieval by Han et al. (1998). Han et al. (1998) obtain a global mean droplet burden of $4 \times 10^{10} \text{ m}^{-2}$, but this value refers to an average over cloudy pixels with liquid cloud tops only ($N_{j,\text{cloudy}}^G$). The analogous calculation for the simulations yields high values of $7.9 \times 10^{10} \text{ m}^{-2}$ (CTL) and $8.5 \times 10^{10} \text{ m}^{-2}$ (AP). The reasons for this overestimation are explored in section 3.5.7.

Global aerosol sources, burdens and lifetimes are given in Table 3.3. For comparison, values from a previous version of ECHAM5-HAM (Stier et al., 2005) and AeroCom all-models averages (Textor et al., 2006) are included. ECHAM5-HAM emissions of sulfate precursors, black carbon and organic carbon are prescribed. Therefore the sulfate, black carbon and organic carbon sources differ only with respect to the conversion of SO$_2$ to sulfate, which depends on atmospheric parameters, between the simulations CTL, AP and Stier et al. (2005). Sea salt and mineral dust emissions depend on the simulated wind field and are both higher in CTL and AP than in Stier et al.’s (2005) study. Differences between our model and ECHAM5-HAM as used by Stier et al. (2005) include the resolution (T42 versus T63), the mode of operation (climatological versus nudged mode), and several differences in the cloud scheme. Stier et al.’s (2005) simulation was run with a single-moment cloud scheme (Lohmann and Roeckner, 1996) and with a statistical cloud cover parameterization (Tompkins, 2002). The simulations presented here include double-moment cloud microphysics (Lohmann et al., 2007), and the cloud cover calculation is based on relative humidity (Sundqvist et al., 1989). It has been shown in a study with the ECHAM4 GCM that simulated mineral dust emissions in the climatological mode are about 25% higher than if the model is nudged to wind fields from a reanalysis (Timmreck and Schulz, 2004), a possible explanation for the difference in our simulations and Stier et al. (2005). It is not clear why mineral dust and sea salt emissions in AP are higher than in CTL, but surface winds over arid regions can be increased in the model due to aerosol feedbacks on clouds, leading to a different model “weather”. Simulations covering
3.4 In-cloud aerosol budgets

Differences in the aerosol lifetime, burden, the aerosol optical depth, and consequently in the cloud droplet concentrations and further cloud parameters are caused by the different treatment of in-cloud aerosol in simulations CTL and AP. Of highest impact are differences in the wet removal of particles from the atmosphere.

3.4.1 Scavenged aerosol mass

The scavenged fraction, i.e., the fraction of aerosol mass and number which is incorporated in hydrometeors and is removed from the atmosphere when precipitation forms, is prescribed to fixed values for the seven modes and for three temperature ranges in the ECHAM5-HAM standard version (Stier et al., 2005). For stratiform liquid clouds, these parameters range from 0.1 for the nucleation mode to 0.99 for the mixed coarse mode (see Table 3.4). With the prognostic treatment of in-cloud particles, the scavenged mass depends on the history of the cloud (vertical velocities at cloud base, time available for collision scavenging, Bergeron-Findeisen process).

Figure 3.2 compares the scavenged aerosol mass simulation CTL, diagnosed with the fixed scavenging parameters, to the prognostic in-droplet and in-crystal mass in simulation AP. The scavenged aerosol masses are in general smaller in simulation AP than in CTL, except at high altitudes. In the ice-cloud levels above approximately 400 hPa, most of the available aerosol mass is in-cloud in simulation AP, because the large soluble aerosol particles are assumed to freeze homogeneously (Hoose et al., 2008b; Kärcher and Lohmann, 2002). In contrast, in CTL, the scavenged mass fraction in ice clouds is assumed to be only 0.1 for all modes.

At lower levels, the scavenged mass is similar between the two simulations for sulfate. For black carbon and organic carbon, the scavenged mass is considerably smaller in simulation AP, because the carbon particles are generally small and therefore rarely activate to cloud droplets. Collision scavenging, though of some importance at these particle sizes, can not compensate for the low nucleation scavenging. For mineral dust, about half of the mass is in the insoluble modes, which are not assumed to activate to cloud droplets at all (Hoose et al., 2008b; Lohmann, 2007), and collision scavenging is negligible for the coarse modes. In simulation CTL, on the other hand, 40% of the insoluble dust is assumed to be scavenged in clouds at temperatures warmer than −35°C. Therefore the scavenged mass is lower approximately by a
Table 3.3: Global annual mean sources, burden and lifetimes in simulations CTL and AP and previous studies. The components are SO$_4$ = (condensed) sulfate, SO$_4$(g) = gaseous sulfuric acid, SO$_2$ = gaseous sulfur dioxide, BC = black carbon, OC = organic carbon, SS = sea salt, DU = dust. The abbreviation of the modes are: NS = nucleation soluble, KS = Aitken soluble, AS = accumulation soluble, CS = coarse soluble, KI = Aitken insoluble, AI = accumulation insoluble, CI = coarse insoluble.

<table>
<thead>
<tr>
<th>Component</th>
<th>CTL</th>
<th>AP</th>
<th>ECHAM5-HAM (Stier et al., 2005)</th>
<th>AeroCom all-models mean (Textor et al., 2006)</th>
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</thead>
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<td>75.9</td>
<td>75.7</td>
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<td>66.1</td>
<td>66.1</td>
<td>96.6</td>
</tr>
<tr>
<td>SS sources</td>
<td>6556</td>
<td>6617</td>
<td>5032</td>
<td>16600</td>
</tr>
<tr>
<td>DU sources</td>
<td>704</td>
<td>869</td>
<td>662</td>
<td>1840</td>
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<td>0.11</td>
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</tr>
<tr>
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<td>0.96</td>
<td>0.99</td>
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<tr>
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<td>13.3</td>
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<tr>
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<td>OC lifetime</td>
<td>5.8</td>
<td>5.3</td>
<td>5.4</td>
<td>6.4</td>
</tr>
<tr>
<td>SS lifetime</td>
<td>0.7</td>
<td>0.7</td>
<td>0.8</td>
<td>0.2</td>
</tr>
<tr>
<td>DU lifetime</td>
<td>4.3</td>
<td>5.2</td>
<td>4.6</td>
<td>3.8</td>
</tr>
</tbody>
</table>

Table 3.4: Scavenging coefficients $R_j$ for stratiform clouds, applied to both mass and number, of the seven modes in standard ECHAM5-HAM (simulation CTL). Adapted from Stier et al. (2005). The abbreviations of the modes are listed in Table 3.3.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>$R_{NS}$</th>
<th>$R_{KS}$</th>
<th>$R_{AS}$</th>
<th>$R_{CS}$</th>
<th>$R_{KI}$</th>
<th>$R_{AI}$</th>
<th>$R_{CI}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0^\circ$C &lt; $T$</td>
<td>0.10</td>
<td>0.25</td>
<td>0.85</td>
<td>0.99</td>
<td>0.20</td>
<td>0.40</td>
<td>0.40</td>
</tr>
<tr>
<td>$-35^\circ$C &lt; $T$ &lt; $0^\circ$C</td>
<td>0.10</td>
<td>0.40</td>
<td>0.75</td>
<td>0.75</td>
<td>0.10</td>
<td>0.40</td>
<td>0.40</td>
</tr>
<tr>
<td>$T$ &lt; $-35^\circ$C</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
</tr>
</tbody>
</table>
Figure 3.2: Zonal and annual mean scavenged aerosol mass, diagnosed from simulation CTL (left), and simulated in simulation AP (right), in $\mu g \ m^{-3}$. 

Figures (left): Scavenged mass SO$_4$ - CTL, Scavenged mass BC - CTL, Scavenged mass OC - CTL, Scavenged mass SS - CTL, Scavenged mass DU - CTL.
Figures (right): Scavenged mass SO$_4$ - AP, Scavenged mass BC - AP, Scavenged mass OC - AP, Scavenged mass SS - AP, Scavenged mass DU - AP.
factor of 5 with the prognostic treatment in simulation AP. This results in an increase of more than 50% of the insoluble accumulation and coarse mode particle number burdens (Table 3.3).

3.4.2 Transfer rates between in-cloud and interstitial modes

Figure 3.3 illustrates the source and sink terms for aerosol in the interstitial, in-droplet and in-crystal modes, calculated from simulation AP. Sulfate differs from the other aerosol components in that it is mainly formed in the atmosphere and not emitted as primary particles. It can either nucleate directly from the gas phase (negligible for the mass budget, but important for particle numbers), condense on pre-existing interstitial particles, or form by heterogeneous reactions in the aqueous phase, in which case it is attributed to the in-droplet mode. The in-cloud formation is the main source (68% of the total production) of atmospheric sulfate. Therefore, release of particles from evaporating hydrometeors is the most important source of interstitial sulfate (Figure 3.3, upper left plot). For black carbon and organic carbon, release during evaporation is roughly as important as the primary emissions. However, these two processes occur at different altitudes: within the atmosphere (release from evaporating hydrometeors) versus only in the two lowest model layers (emissions). The source term at higher altitudes can have a bigger impact, because aerosol mass concentrations and their sinks generally decrease with altitude. Sea salt and dust are emitted from the surface as large particles. Sedimentation is the dominating removal process and can eliminate coarse-mode particles after a short lifetime before interaction with clouds. The other interstitial particles are more likely to be removed from the interstitial phase when cloud droplets or ice crystals nucleate on them, or by below-cloud and convective wet deposition. Collisions with cloud droplets or ice crystals are only significant for black carbon and organic carbon, which constitute the insoluble Aitken mode. The different relative contributions from in-cloud sulfate formation, droplet nucleation and collision scavenging are also obvious in the middle column of Figure 3.3. Furthermore, it is also noticeable that the contributions of the sink terms for in-droplet particles vary. Sea salt and sulfate aerosols are removed by wet deposition with a higher likelihood than black carbon, organic carbon and dust. This is related to the origins of the clouds into which the particles are incorporated; e.g. clouds in a marine environment, with fewer droplets, can form precipitation faster than polluted continental clouds.

Aerosol mass is incorporated into in-crystal particles largely by ice nucleation (Figure 3.3, right column), and removed by precipitation formation. Freezing of cloud droplets contributes to the in-crystal aerosol mass only in a negligible amount. Only a low number of droplets freezes in mixed-phase clouds, while the majority subsequently evaporates during the Wegener-Bergeron-Findeisen process. Again, collision scavenging is relevant only for the smaller particles sulfate, black carbon and organic carbon.

3.4.3 Life cycles of cloud condensate and in-cloud aerosol

In this section we compare the time scales of processing of water vapor and aerosols by clouds, as simulated by ECHAM5-HAM with the above-described aerosol processing scheme, to back-of-the-envelope calculations by Pruppacher and Jaenicke (1995). Pruppacher and Jaenicke (1995) based their estimates on global mean values of cloud parameters (cloud cover, liquid water content, vertical velocities and others) for different cloud types. Selected results are summarized in Table 3.5. Their estimate of the global mean liquid water path (LWP) is very high with 388 g m$^{-2}$. Current estimates from satellite retrievals over ocean range between 50 – 84 g m$^{-2}$ (Lohmann et al., 2007). E.g. Bennartz (2007) show that the liquid water path over ocean, derived from Advanced Scanning Microwave Radiometer (AMSR) measurements, rarely exceeds 200 g m$^{-2}$. On the other hand, the estimate of global mean precipitation ($P$) to 1000 mm yr$^{-1}$ is in accordance with observations from the Global Precipitation Climatology Project (Adler et al., 2003). Along with the high LWP, Pruppacher and Jaenicke (1995) estimate a high condensation rate $C_{c,c}$ of about 11 times the precipitation rate. The evaporation rate $E_{v,c}$ is in between condensation and (surface) precipitation. It thus includes below-cloud evaporation. According to Pruppacher and Jaenicke (1995) more than 90% of the condensed cloud water reevaporates, rather than forming precipitation which reaches the ground. Therefore the cloud lifetime with respect to precipitation ($\tau_{c,precip} = 3.4$ h) is ten times as long as the cloud lifetime with respect to evaporation ($\tau_{c,\text{evap}} = 20.5$ min).

As Pruppacher and Jaenicke (1995) give the cloud parameters for 5 cloud types separately, we can exclude the convective clouds Cb (cumulonimbus) and Cu (cumulus) from the analysis and recalculate the cloud condensate budget for stratiform clouds only. We assume that 50% of the global precipitation originates from stratiform clouds (based on Tropical Rainfall Measuring Mission (TRMM) satellite precipitation radar observations between 40$^\circ$S and 40$^\circ$N, reported by Tost et al. (2006)). For stratiform clouds only 72% of the condensate reevaporates. The lifetime with respect to precipitation is shorter
Figure 3.3: Annual zonal mean transfer rates for interstitial (left), in-droplet (middle row) and in-crystal (right) aerosol mass, integrated vertically over the atmospheric column. The legend in the upper plots refers to all plots in the respective column. Contributions from freezing and melting are negligible. Note the different scales.
Table 3.5: Global budgets of cloud water and aerosols. P&J stands for Pruppacher and Jaenicke (1995). LWP: liquid water path, TWP: total water path (given in the last column only), $P$: precipitation, $C_{v,c}$: rate of condensation of water vapor mass, $E_{v,in-cloud}$: rate of in-cloud evaporation of water mass, $E_{v,c}$: rate of in-cloud evaporation of cloud water mass plus below-cloud evaporation of precipitation, $\tau_{c,\text{precip}}$: cloud lifetime with respect to precipitation, $\tau_{c,\text{evap}}$: cloud lifetime with respect to evaporation, $C_{\text{AP},c}$: rate of aerosol mass transferred into clouds, $E_{\text{AP},c}$: rate of aerosol mass released from evaporating clouds, $W_{\text{dep},ic}$: in-cloud wet deposition of aerosol mass, $W_{\text{dep},bc}$: below-cloud wet deposition of aerosol mass, $W_{\text{dep}}$: total wet deposition of aerosol mass, $S_{\text{AP}}$: source strength of aerosols on the ground.

<table>
<thead>
<tr>
<th></th>
<th>P&amp;J, convective and stratiform clouds</th>
<th>P&amp;J, recalculated for stratiform clouds only</th>
<th>P&amp;J, rescaled LWP</th>
<th>AP, stratiform liquid clouds</th>
<th>AP, stratiform liquid, mixed-phase and ice clouds</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWP (TWP) in gm$^{-2}$</td>
<td>388</td>
<td>136</td>
<td>60</td>
<td>38</td>
<td>60</td>
</tr>
<tr>
<td>$P$ in mm yr$^{-1}$</td>
<td>1000</td>
<td>500</td>
<td>500</td>
<td>78$^a$</td>
<td>486</td>
</tr>
<tr>
<td>$P$ in g m$^{-2}$ h$^{-1}$</td>
<td>114</td>
<td>57</td>
<td>57</td>
<td>8.9$^a$</td>
<td>55.5</td>
</tr>
<tr>
<td>$C_{v,c}$ in g m$^{-2}$ h$^{-1}$</td>
<td>1256</td>
<td>206</td>
<td>90</td>
<td>95.4</td>
<td>156.7</td>
</tr>
<tr>
<td>$E_{v,in-cloud}$ in g m$^{-2}$ h$^{-1}$</td>
<td>1142</td>
<td>148</td>
<td>33</td>
<td>86.4$^a$</td>
<td>101.2</td>
</tr>
<tr>
<td>$E_{v,c}$ in g m$^{-2}$ h$^{-1}$</td>
<td>0.91</td>
<td>0.72</td>
<td>0.37</td>
<td>0.91</td>
<td>0.65</td>
</tr>
<tr>
<td>$\tau_{c,\text{precip}} = \frac{\text{LWP}}{P}$ in h</td>
<td>3.4</td>
<td>2.4</td>
<td>1.6</td>
<td>4.2</td>
<td>1.1</td>
</tr>
<tr>
<td>$\tau_{c,\text{evap}} = \frac{\text{LWP}}{E_{v,c}}$ in min</td>
<td>21</td>
<td>55</td>
<td>110</td>
<td>26</td>
<td>36</td>
</tr>
<tr>
<td>$C_{\text{AP},c}$ in Tg yr$^{-1}$</td>
<td>6533</td>
<td>1072</td>
<td>473</td>
<td>3287</td>
<td>4503</td>
</tr>
<tr>
<td>$E_{\text{AP},c}$ in Tg yr$^{-1}$</td>
<td>5947</td>
<td>775</td>
<td>171</td>
<td>2925$^b$</td>
<td>3395</td>
</tr>
<tr>
<td>$W_{\text{dep},ic}$ in Tg yr$^{-1}$</td>
<td>586</td>
<td>297</td>
<td>302</td>
<td>1950$^c$</td>
<td>2952$^c$</td>
</tr>
<tr>
<td>$W_{\text{dep},bc}$ in Tg yr$^{-1}$</td>
<td>146</td>
<td>74</td>
<td>76</td>
<td>1694$^{c,d}$</td>
<td>1694$^{c}$</td>
</tr>
<tr>
<td>$W_{\text{dep}}$ in Tg yr$^{-1}$</td>
<td>732</td>
<td>371</td>
<td>378</td>
<td>2285</td>
<td>2790</td>
</tr>
<tr>
<td>$S_{\text{AP}}$ in Tg yr$^{-1}$</td>
<td>$\approx$ 2000</td>
<td>2000</td>
<td>2000</td>
<td>7851</td>
<td>7851</td>
</tr>
<tr>
<td>$E_{\text{AP},c}$</td>
<td>0.91</td>
<td>0.72</td>
<td>0.37</td>
<td>0.89</td>
<td>0.75</td>
</tr>
<tr>
<td>$E_{\text{AP},c}$</td>
<td>$\approx$ 3</td>
<td>$\approx$ 0.4</td>
<td>0.09</td>
<td>0.37</td>
<td>0.43</td>
</tr>
</tbody>
</table>

$^a$ under the assumption of a 40% contribution to evaporation of cloud condensate by below-cloud evaporation (which has been diagnosed for all stratiform clouds)

$^b$ under the assumption of a 55% contribution to release of aerosol mass from evaporating droplets by below-cloud evaporation (which has been diagnosed for all stratiform clouds)

$^c$ before below-cloud evaporation of precipitation

$^d$ includes below-cloud scavenging through precipitation from mixed-phase/ice clouds

$^e$ after below-cloud evaporation
(\(\tau_{\text{precip}} = 2.4\) h) and the lifetime with respect to evaporation is longer (\(\tau_{\text{evap}} = 55\) min). In the third column of Table 3.5, the liquid water path of Pruppacher and Jaenicke’s (1995) estimate for stratiform clouds is scaled down to a more realistic value of 60 g m\(^{-2}\). The condensation rates are changed accordingly, but the value of stratiform precipitation is kept constant, because it agrees well with observations. The evaporation rate \(E_{\text{v,c}}\) can subsequently be calculated from the difference between condensation and evaporation. The resulting cloud lifetime with respect to precipitation reduces to 1.6 h.

ECHAM5-HAM (simulation AP) simulates a substantially lower global mean LWP of stratiform clouds of 38 g m\(^{-2}\). 35% of the modelled global mean precipitation is stratiform. The fraction of this precipitation originating from liquid clouds can only be estimated indirectly, because the diagnostics of below-cloud evaporation does not distinguish between precipitation originating from liquid or mixed-phase/ice clouds. The in-cloud evaporation rate \(E_{\text{v,in-cloud}}\) is diagnosed for liquid and ice separately. For all stratiform clouds, \(E_{\text{v,c}}\) can be calculated from \(C_{\text{v,c}} - P\). With this we obtain \(E_{\text{v,in-cloud}}/E_{\text{v,c}} = 60\%\) for all stratiform clouds and apply the same ratio to liquid clouds only, to finally calculate \(P\) from liquid clouds. We conclude that only a small fraction of precipitation from liquid clouds (20%) reaches the ground. Similarly high fractions (80–85%) of evaporating precipitation have been found in field studies and simulations of drizzle from stratuscumulus clouds (Bretherton et al., 2004; Wood, 2005a). The condensation rate \(C_{\text{v,c}}\) is 11 times as large as the precipitation rate \(P\), which leaves 91% of the cloud condensate to re-evaporate. The simulated lifetime with respect to precipitation is \(\tau_{\text{precip}} = 4.2\) h, and the lifetime with respect to evaporation is \(\tau_{\text{evap}} = 44\) min. When mixed-phase and ice clouds are also taken into account, the total water path amounts to 60 g m\(^{-2}\). Mixed-phase/ice clouds tend to precipitate more frequently. Therefore the lifetime with respect to precipitation for all stratiform clouds in simulation AP is 1.1 h, and the lifetime with respect to evaporation is 36 min.

For an estimation of the aerosol cycling through clouds, Pruppacher and Jaenicke (1995) assume that aerosol particles are incorporated into clouds and released from them in the same way as water molecules. Furthermore, their calculations are based on a mean aerosol mass concentration of 1 \(\mu g m^{-3}\), and a scavenging efficiency of 1, i. e. all aerosol particles inside clouds are assumed to be incorporated into hydrometers. The rate of aerosol mass transferred into clouds, \(C_{\text{AP,c}}\), additionally contains a 30% contribution of aerosol material converted from gases which have dissolved in the cloud droplets. As for cloud water, the fraction of aerosol particles released from evaporating droplets (\(E_{\text{AP,c}}\)) is 91%, and only 9% is removed from the atmosphere by in-cloud wet deposition \(W_{\text{dep,ic}}\). Below-cloud wet deposition \(W_{\text{dep,bc}}\) is assumed to contribute to 20% to the total aerosol wet deposition. Comparing the rate of aerosol release from evaporating clouds to the aerosol surface source \(S_{\text{AP}}\), which is estimated to \(\approx 2000\) Tg yr\(^{-1}\), yields the number of aerosol cycles through clouds: \(E_{\text{AP,c}}/S_{\text{AP}} \approx 3\). Repeating the calculations for stratiform clouds only gives a fraction of particles released during droplet evaporation of 72% (same as for cloud condensate). The average number of cycles through stratiform clouds is only approximately 0.4. Continuing the downscaling of the global liquid water path to a more realistic value for the uptake of aerosols into clouds, finally yields 0.09 aerosol cycles through clouds. This implies that only every tenth aerosol undergoes a cycle through a stratiform cloud. The aerosol emission strength has not been modified, although it seems to be too low compared to current model estimates. The highest contribution to the aerosol mass emissions is sea salt (Table 3.3), but for a marine boundary layer environment the average aerosol concentration of 1 \(\mu g m^{-3}\) as assumed by Pruppacher and Jaenicke (1995) is an order of magnitude too low (see also section 3.5.2 below). The same argumentation applies for mineral dust. Therefore this estimate can be seen as valid for average accumulation mode aerosols, without a noticeable coarse mode fraction.

In ECHAM5-HAM, the surface source of aerosol mass is 4 times as large as estimated by Pruppacher and Jaenicke (1995). Therefore the transfer rates of aerosol mass into clouds, the release rates from evaporating clouds and the wet deposition rates are all considerably larger. In-cloud and below-cloud wet deposition can not be diagnosed with the separate contributions from below-cloud evaporation. Therefore the sum of \(W_{\text{dep,ic}}\) and \(W_{\text{dep,bc}}\) is larger than \(W_{\text{dep}}\), and the difference gives the contribution to \(E_{\text{AP,c}}\) by evaporating rain or snow particles. Different from evaporating clouds, for evaporating precipitation an inhomogeneous mixing assumption is applied. This implies that if a certain fraction of precipitation mass evaporates in a dry below-cloud layer, the same fraction of in-rain/in-snow aerosol mass and number is released. Below-cloud evaporation contributes to 55% to the total release of aerosols from evaporating water condensate, which is higher than for cloud condensate (40%), because inside clouds a homogeneous mixing assumption is used. I. e. when cloud water evaporates without a change in the cloud fraction, it is assumed that all cloud droplets shrink by the same amount, but their number is not reduced. Therefore during this process no aerosol particles are released, contrarily to the assumption which is underlying Pruppacher and Jaenicke’s (1995) calculations. Due to these different mixing assumptions, the ratio of
evaporation over condensation for aerosols \((E_{AP,c}/C_{AP,c} = 0.89\) and 0.75, for only liquid and for all stratiform clouds, respectively), is not the same as for cloud condensate \((0.91/0.65)\). Together with the high surface source term, the ratio of \(E_{AP,c}/S_{AP}\), which is the mean number of cycles through stratiform clouds experienced by an aerosol particle between its emission from the surface and its deposition, is 0.37 for liquid clouds and 0.43 for all stratiform clouds.

The dimensionless values \(E_{c,c}/C_{c,c}, E_{AP,c}/C_{AP,c}\) and \(E_{AP,c}/S_{AP}\) obtained for all stratiform clouds in simulation AP are similar to Pruppacher and Jaenicke (1995), recalculated for stratiform clouds, although their original estimates of the liquid water path and of the aerosol emission are not realistic. Rescaling Pruppacher and Jaenicke’s (1995) liquid water path yields a significantly lower amount of cloud cycles. The concept of calculations based on global mean values is doubtful for highly heterogeneous fields like aerosol concentrations. Here model simulations can provide more accurate estimates. On the other hand, it has become obvious that aerosol cycling through convective clouds contributes significantly to global aerosol processing, and an extension of the model in this direction is desirable. Furthermore, as below-cloud evaporation contributes significantly to the release of aerosol mass from hydrometeors, a better treatment of this process is necessary. Here, a prognostic double-moment formulation for rain (recently implemented by Posselt and Lohmann, 2007) is necessary to accurately calculate the size of aerosol particles released in dry layers below clouds.

### 3.5 Comparison with observations

Comparisons between global climate model simulations and observations are hampered by the different scales in space and time for which simulated and observed values are representative. If a GCM is not nudged to the synoptic conditions at the observation time, the model results can only be compared as climatological mean values to long timeseries of observations. Furthermore, many observations reflect local conditions, which can vary within a few kilometers, while a GCM gridbox size in T42 resolution is over 300 km x 300 km at the equator. Here we have chosen several observations of aerosol or cloud microphysical parameters, which cover either large parts of the globe (sections 3.5.1 –3.5.5) or are based on the statistical analysis of clouds sampled under different conditions at one location (section 3.5.6).

#### 3.5.1 Marine boundary layer aerosol

Heintzenberg et al. (2000) provide a compilation of aerosol concentration and size distribution measurements in the marine boundary layer (MBL). A multimodal lognormal size distribution was fitted to the original data, and from this the number concentrations and dry mean diameters of the Aitken and accumulation modes are given as zonal mean values. These observations are compared to the ECHAM5-HAM simulations CTL and AP in Figures 3.4 and 3.5. The model data were averaged over the surface level at all ocean gridpoints.

While the Aitken mode zonal mean number concentrations are slightly lower in AP than in CTL, the accumulation mode number concentrations are significantly higher. The Aitken mode concentrations in both simulations are reasonably close to observations on the Northern Hemisphere, but too low between 15 and 60°S. The accumulation mode numbers in simulation AP agree better with the observations. Both the Aitken and the accumulation mode zonal mean diameters are similar in CTL and AP, but smaller in AP. For the Aitken mode CTL agrees slightly better with the observations on the Southern Hemisphere. The accumulation mode diameter is overestimated up to 100% by both simulations, especially at higher latitudes. Furthermore, an increase towards the south is simulated, while the observations are relatively constant or decrease slightly. The diameter overestimation is probably due to the size of the emitted sea salt particles in ECHAM5-HAM. The windspeed-dependent mass median radii of accumulation mode sea salt particles ranges between 271 and 284 nm (Stier et al., 2005). Conversion to number mean diameters gives 317 to 332 nm. As sea salt is expected to be the dominant aerosol type in remote ocean regions, the average number mean diameter in the model is probably dominated by this prescribed value. In the light of the observed diameters (Heintzenberg et al., 2000), which are on average 140 nm at high Southern latitudes, the emitted particle size might be too large. Figure 3.5 illustrates the marine aerosol size distributions and reflects the improvement in the accumulation mode numbers.

#### 3.5.2 Sea salt mass concentrations

Figure 3.6 compares sea salt mass concentrations in the lowest model level to long-term measurements from the North American IMPROVE network and remote marine sites provided by J. Prospero and
3.5 Comparison with observations

Figure 3.4: Aitken and accumulation mode number concentrations and number mean diameters over the oceans, averaged over zonal bands, in simulations CTL and AP, compared to observations compiled by Heintzenberg et al. (2000).

Figure 3.5: Size distributions over the oceans, averaged over zonal bands, resulting from the parameters in Figure 3.4, from simulations CTL, AP and observations compiled by Heintzenberg et al. (2000).
The Aerosol Robotic Network (AERONET) provides ground-based remote sensing observations of aerosol optical parameters from a large world-wide network of automated sun photometers (Holben et al., 1998). AERONET results from 1993–2003 have been used to derive column mean aerosol size distributions and aerosol volume burdens for 103 stations (G. Lesins, personal communication). The same dataset has been used by Ma and von Salzen (2006) to evaluate simulations of sulfate aerosol size distributions.

The column integrated aerosol volume size distribution \( \frac{dV(d)}{d \ln d} \) has been calculated for simulations CTL and AP as follows, as a function of the particle diameter \( d \).

\[
\frac{dV(d)}{d \ln d} = \sum_k \left( \sum_{j=1}^{7} \frac{\pi}{6} d^2 \exp \left(-\frac{(d - 2r_{\text{wet},j,k})^2}{2 \ln^2 \sigma_j}\right) \right) \frac{\Delta p_k}{g \rho_{\text{air},k}}
\]

The index \( k \) runs over all vertical levels. \( N_{j,k} \) is the aerosol number concentration of mode \( j \) in level \( k \), and \( r_{\text{wet},j,k} \) the median aerosol wet radius of mode \( j \) in level \( k \). The standard deviation \( \sigma_j \) is fixed to the value of 2.0 for the coarse modes and to 1.59 for all other modes. The vertical integral is weighted with the geometrical layer thicknesses, calculated from the pressure difference \( \Delta p_k \) between adjacent layer interfaces, the acceleration of gravity \( g \) and the air density \( \rho_{\text{air},k} \). As the sun photometers only measure during cloud-free conditions, the simulated data are filtered to include only points with a cloud fraction smaller than 15\% (similar to the analysis by Ma and von Salzen (2006)). The AERONET size distributions are provided for the radius range of 50 nm to 15 \( \mu \)m, i.e. the accumulation, coarse and supercoarse modes.

In Figure 3.7, the size distributions for all stations calculated by equation (3.5) and the total volume, integrated over the size distribution, from simulations CTL and AP are compared to the AERONET retrievals. The simulated size distributions have a higher variance than the AERONET size distributions. The accumulation mode wet diameter is frequently overestimated in both simulations. Simulation AP exhibits higher particle numbers in the nucleation and Aitken modes \( (d < 0.1 \mu \text{m}) \), where no observations are available. This is consistent with the higher nucleation and Aitken mode global number burdens (compare Table 3.3). The total vertically integrated aerosol volume correlates poorly between both simulations and the observations. In Figure 3.9, the analysis is split into different regions, as mapped in Figure 3.8. In all continental regions, the total volume is generally underestimated except for some European and South American stations. On all ocean stations, on the other hand, ECHAM5-HAM overestimates the total volume by up to an order of magnitude. Consistent with Figure 3.4 and the conclusion of section 3.5.1 that the emitted size of sea salt particles in ECHAM5-HAM might be too large, the mismatch in the accumulation mode diameter is also worst for the ocean stations. A second reason for the overestimation of the volume can also be the aerosol water uptake, as here aerosol size distributions at ambient relative humidity are compared. The AeroCom model intercomparison (Textor et al., 2006) has revealed that except for one outlier, ECHAM5-HAM has the highest global water uptake by aerosols relative to the aerosol dry mass. This is related to the fact that unlike in most other models, the sea salt burden in ECHAM5-HAM is higher than the dust burden, and the hygroscopic salt particles take up more water.

On a number of stations, AERONET measured annual mean volume burdens of more than 0.15 \( \mu \text{m}^3 \mu \text{m}^{-2} \). These are mainly stations in arid regions, especially in West Africa and at the Persian Gulf. The high values can possibly be influenced by severe dust events, and thus are not reproduced by the model. Also at two stations in South America, numbers 40 and 52 in Figure 3.8, the model underestimates the observed elevated volume burdens.
3.5 Comparison with observations

Figure 3.6: Sea salt (SS) mass concentrations at University of Miami and IMPROVE stations. The upper plot displays the data obtained with a previous version of ECHAM5-HAM by Stier et al. (2005). The lower row shows a comparison of simulation CTL versus simulation AP for total sea salt mass and sea salt mass in the accumulation mode.
Figure 3.7: Vertically integrated size distributions and volume burden for all analyzed AERONET stations (G. Lesins, personal communication) compared to simulations CTL and AP.

Figure 3.8: AERONET stations used for Figures 3.7 and 3.9. Red: Eastern US, magenta: Europe, green: Western US, gray: South America, blue: ocean, black: other stations (G. Lesins, personal communication).
3.5 Comparison with observations

Figure 3.9: Vertically integrated size distributions and volume burden for AERONET stations (G. Lesins, personal communication) from five different regions, see Figure 3.8, compared to simulations CTL and AP.
3.5.4 AERONET aerosol optical depth

At the AERONET stations listed in Figure 3.8, the aerosol optical depth (AOD) and absorption aerosol optical depth (ABS) were measured, too. The aerosol optical depth is defined as the vertical integral from the surface to the top of the atmosphere (TOA) over the aerosol extinction coefficient \( b_{\text{ext}} \), which includes both scattering \( b_{\text{scat}} \) and absorption \( b_{\text{abs}} \) of radiation. \( b_{\text{scat}} \) and \( b_{\text{abs}} \) depend on the wavelength \( \lambda \), the particle concentration and size distribution in the respective layer, and on the particles’ complex refractive index. The absorption aerosol optical depth is defined as the vertical integral over \( b_{\text{abs}} \) (Seinfeld and Pandis, 1998).

\[
\text{AOD} (\lambda) = \int_{z=0}^{\text{TOA}} b_{\text{ext}} (\lambda, z) \, dz = \int_{z=0}^{\text{TOA}} (b_{\text{scat}} (\lambda, z) + b_{\text{abs}} (\lambda, z)) \, dz
\]  
\[
\text{ABS} (\lambda) = \int_{z=0}^{\text{TOA}} b_{\text{abs}} (\lambda, z) \, dz
\]  

From measurements at 500, 670 and 870 nm (AOD) and 441 and 673 nm (ABS) wavelength, the data have been linearly interpolated to 550 and 825 nm for comparison with the simulations. The results are shown in Figure 3.10. The correlation with the observations is rather poor for both simulations. Simulation AP simulates excessively high values of the AOD over ocean. In a sensitivity study (simulation AP-aod) it has been tested to which extent this is due to the swelling of sea salt aerosols. The water uptake of salt-containing particles has been reduced by a factor of 0.5. Figures 3.10(c) and (g) illustrate that this reduces the AOD at the gridpoints with the highest simulated values by approximately 30%. The global mean AOD at 550 nm decreases from 0.35 to 0.25, but this is still higher than the retrieval by S. Kinne (personal communication) of 0.15 – 0.19 (see Table 3.2).

Absorption aerosol optical depth (ABS), depicted in Figures 3.10(h), (i) and (j), is underestimated at most stations by all three simulations. Stier et al. (2007) noted the same for his BASE simulation (similar to Stier et al. (2005)) when comparing to a different set of AERONET retrievals. The simulated global mean values of ABS are \( 2.1 \times 10^{-3} \) in simulation CTL, same as in Stier et al.’s (2007) BASE simulation, and \( 2.0 \times 10^{-3} \) (\( 1.8 \times 10^{-3} \)) in simulation AP (AP-aod). Stier et al. (2007) showed that using recent estimates of the refractive index for black carbon by Bond and Bergstrom (2006) increases the global mean ABS to \( 3.0 \times 10^{-3} \) and improves the correlation to AERONET measurements.

3.5.5 Marine boundary layer cloud droplets

As the number of MBL accumulation mode particles changes significantly with the new treatment of in-cloud particles, this also has a substantial influence on MBL clouds. Droplet number concentrations in marine boundary layer clouds are retrieved from MODIS satellite data by Bennartz (2007). The dataset has been extended to 5.25 years (R. Bennartz, personal communication) and covers nearly the complete ocean region between 60°N and 60°S on a \( 1° \times 1° \) grid. The in-cloud droplet concentration is retrieved on a daily basis. Only gridpoints with more than 10 days with a low cloud fraction higher than 80% are considered in the analysis. The average over all stratiform liquid cloud cases in the observation period is depicted in Figure 3.11. This Figure also displays the in-cloud droplet concentrations \( N_l \) in simulations CTL and AP as an annual average over the lowest 4 model layers (approximately 900 m). Simulation AP yields significantly higher droplet concentrations than simulation CTL all over the globe. Compared to Bennartz (2007), the pattern of high concentrations along the coasts of North America and east Asia, in the Gulf of Mexico and in the Mediterranean Sea are qualitatively well captured in both simulations. In other near-coast regions the model results and satellite retrievals disagree. The observed enhanced concentrations along the Peruvian and southern African west coasts, which are stratocumulus regions, are lacking in both simulations. On the other hand the droplet concentrations are overestimated in the tropical Atlantic, in the northern Indian ocean and between Australia and Indonesia, where the observations do not show higher than average values. In general the cloud droplet concentration fields are more homogeneous in the model than in the observed data. The observations yield minimum values below 25 cm\(^{-3}\) in the remote oceans, suggesting that the minimum droplet concentration of 40 cm\(^{-3}\) in ECHAM5-HAM (Lohmann et al., 2007, based on typical concentration ranges given by Quante (2004)) is too high and should be reconsidered.

A more quantitative comparison to Bennartz’s (2007) retrieval is shown in Figure 3.12. The values are averaged over five selected off-coast regions, some of them dominated by stratocumulus, and five
3.5 Comparison with observations

Figure 3.10: Comparison of aerosol optical thickness (AOD) and absorption (ABS) measurements at AERONET stations (G. Lesins, personal communication) with simulations CTL (circles), AP (triangles) and AP-aod (squares).
Figure 3.11: In-cloud droplet concentrations in marine boundary layer clouds from satellite retrievals by Bennartz (2007) compared to simulations CTL and AP. White: no data.
3.5 Comparison with observations

Figure 3.12: In-cloud droplet concentrations in marine boundary layer clouds in simulations CTL and AP, compared to retrievals by Bennartz (2007) for the regions displayed in Figure 3.13. Error bars indicate one standard deviation. (a) includes averages over all clouds, while for (b) the retrievals are for clouds with a low likelihood of drizzle only.

Figure 3.13: Regions for which the average values in Figure 3.12 are calculated.

remote ocean regions with a distance from land of more than 1500 km (for the regions see Figure 3.13). Bennartz (2007) provides two different estimates for all clouds and for clouds with a low likelihood of drizzle. Droplet concentrations are 17-72% higher for non-drizzling clouds. In ECHAM5-HAM, drizzle is not represented, as cloud droplet which have grown large enough to sediment are assumed to reach the ground within the same timestep. Simulation CTL agrees well with the all-cloud observations, but underestimates the droplet number concentrations when comparing the simulations to non-drizzling clouds only. Simulation AP overestimates the droplet number concentrations by more than a factor of two, with a slightly better agreement for non-drizzling clouds. This is surprising, because the higher droplet concentrations in simulation AP are the result of higher aerosol number concentrations in the marine boundary layer, especially in the accumulation mode, which agree much better with observations in simulation AP (section 3.5.1). Furthermore, compared to the retrieval of vertically integrated droplet concentrations by Han et al. (1998), both simulations are significantly too high (Figure 3.1(d)). This suggests either an overestimation of cloud droplet activation or of cloud thicknesses, a too high minimum value for the cloud droplet concentration, or might be caused by inconsistencies between the different observations and retrievals.

3.5.6 Scavenged fraction at Jungfraujoch

Henning et al. (2004) and Verheggen et al. (2007) have analyzed a large set of observations of interstitial and in-cloud aerosol in mixed-phase clouds at the high-altitude research site Jungfraujoch (Swiss Alps). Aerosol size distributions were measured by a Scanning Mobility Particle Sizer (SMPS) behind two different inlets, one sampling interstitial aerosol and one sampling total aerosol (interstitial plus residuals from hydrometeors). From these measurements, a “scavenged particle number fraction” can be defined as
Figure 3.14: Scavenged fraction versus temperature for simulation CTL. The scavenged fraction is calculated in two different ways for (a) and (c) \( F_{\text{CTL,1}} \) and (b) and (d) \( F_{\text{CTL,2}} \), see text. The boxplots (c) and (d) are based on the same data as the plots above. The boxes and whiskers indicate the 10th, 25th, 50th, 75th and 90th percentiles. The mean per temperature interval is marked with an asterisk. The number of data points per temperature interval is given above each box. Observations by Henning et al. (2004) and Verheggen et al. (2007) are included for comparison.

follows (see also equation (2.19)).

\[
F_N = \frac{N_{\text{tot}}(r > 50\text{nm}) - N_{\text{int}}(r > 50\text{nm})}{N_{\text{tot}}(r > 50\text{nm})} \quad (3.8)
\]

\( N_{\text{tot}} \) is the measured total aerosol concentration, and \( N_{\text{int}} \) the interstitial aerosol concentration. The cut-off of 50 nm is chosen because it is the typical dry radius of the smallest activated particles under the orographic conditions of the Jungfraujoch research station (Baltensperger et al., 1998). The studies by Henning et al. (2004) and Verheggen et al. (2007) have shown that the fraction of scavenged particles decreases with decreasing temperatures in the temperature range \(-25^\circ\) to \(0^\circ\)C. The Wegener-Bergeron-Findeisen process is the most likely explanation for this finding. The lower the temperature, the higher is the probability of cloud glaciation and with that the evaporation of cloud droplets, releasing formerly scavenged particles back into the interstitial phase. For temperatures above \(-5^\circ\)C, Verheggen et al. (2007) found a decrease of the scavenged fraction with increasing total aerosol number.

In Figures 3.14 and 3.15, the simulations CTL and AP are compared to Verheggen et al.’s (2007) and Henning et al.’s (2004) observations. The data are sampled over a whole year of instantaneous data which were saved every 12 hours, from the four gridpoints which are closest to the Jungfraujoch, throughout the lowest 9 model layers (approximately five kilometers). This was required in order to obtain a sample which was large enough for the subsequent statistical analyses. Note that the Alpine topography is not well represented on the coarse model grid. The in-cloud values of the cloud droplet concentration \( N_l \) and the ice crystal concentration \( N_i \) have been used. For simulation CTL, in total 1516 points were in clouds with temperatures between \(-20\) and \(+5^\circ\)C, and 2160 data points in simulation AP.

For simulation CTL, the scavenged fraction \( F_N \) is calculated in two different ways. First, \( F_N \) is diagnosed from the hydrometeor concentration and the total aerosol number concentration \( N_{\text{tot}}(r > 50\text{nm}) = N_{\text{AS}} + N_{\text{CS}} + N_{\text{AI}} + N_{\text{CI}} \), where \( N_j \) are the number concentrations in the accumulation soluble (AS), coarse soluble (CS), accumulation insoluble (AI) and coarse insoluble (CI) modes.

\[
F_{\text{CTL,1}} = \max \left( 1, \frac{N_l + N_i}{N_{\text{tot}}(r > 50\text{nm})} \right) \quad (3.9)
\]
Calculated in this way, the temperature dependency of $F_{\text{CTL},1}$ mainly reflects the temperature dependency of the hydrometeor concentration. In Figure 3.14 (a) and (c) we observe a clustering of values of the scavenged fraction at 1 (in liquid clouds, with the droplet concentration equaling or exceeding $N_{\text{tot}}(r > 50\text{nm})$) and, at temperatures below 0°C, at values below 0.1. The low values can be attributed to pure ice clouds, because the minimum droplet concentration in ECHAM5-HAM is 40 cm$^{-3}$ (which would require a total aerosol concentration of 400 cm$^{-3}$ to give a scavenged fraction of 0.1, and such high values of $N_{\text{tot}}(r > 50\text{nm})$ are not observed in the low temperature range). Some data, especially above $-10^\circ\text{C}$, also fall in the intermediate range of $0.4 \leq F_{\text{CTL},1} < 1$. In Figure 3.14 (c) the data point from Figure 3.14 (a) are binned into temperature intervals of 5°C and displayed as boxes, which are limited by the 25th and 75th percentiles. The median, indicated by the middle line, continuously decreases with temperature from 1.0 to 0.15, but the data are widespread. Therefore the decrease with temperature is less pronounced for the mean of the data. Between 0 and $-10^\circ\text{C}$, the simulated mean is close to the observed mean values (Henning et al., 2004; Verheggen et al., 2007), but at lower temperatures the scavenged fraction is overestimated.

Even though this result looks very promising, we have to take into consideration that this is only a diagnostic analysis. Aerosol scavenging in simulation CTL, for the purpose of wet deposition, is calculated with prescribed scavenging coefficients $R_j$ (Stier et al., 2005), listed in Table 3.4. As a second definition of the scavenged fraction in simulation CTL, we calculate the number-weighted mean scavenging coefficient.

$$F_{\text{CTL},2} = \tilde{R} = \sum_j \frac{N_j}{N_{\text{tot}}(r > 50\text{nm})} R_j(T) \quad \text{with} \quad j = \text{AS, CS, AI, CI}$$

$F_{\text{CTL},2}$ is shown in Figure 3.14 (b) and (d). Virtually all values fall on 0.85 above 0°C, and on 0.75 below 0°C. These are the values of the scavenging coefficient for the accumulation soluble mode, which dominates the number concentration of particles larger than 50 nm. The correlation with the observations is therefore poor.

For simulation AP, the scavenged fraction is calculated similar to the measurements.

$$F_{\text{AP}} = \frac{N_1 + N_i}{N_i + N_1 + N_{\text{int}}(r > 50\text{nm})}$$

By definition, $F_{\text{AP}}$ is always smaller than 1 in clouds. Figure 3.15 shows that similar to Figure 3.14(a), we observe a clustering of values from pure ice clouds below 0.1, and clouds containing liquid with higher scavenged fractions. For AP, the values are lower than for CTL, and the median is around 0.5. As the fraction of liquid clouds decreases with decreasing temperature, the scavenged fraction also exhibits a decreasing trend, similar to but weaker than the observations. The mean scavenged fraction is lower than observed at temperatures above $-5^\circ\text{C}$ and higher than observed at temperatures below $-10^\circ\text{C}$.

Verheggen et al. (2007) furthermore report that $F_N$ decreases monotonically as a function of the total aerosol number concentration $N_{\text{tot}}(r > 50\text{nm})$ for $T > -5^\circ\text{C}$, but is rather insensitive to $N_{\text{tot}}(r > 50\text{nm})$ in the mixed-phase clouds below $-5^\circ\text{C}$. Above $-5^\circ\text{C}$, they find the mean scavenged fraction to decrease from 0.8 to 0.3, while below $-5^\circ\text{C}$, the mean scavenged fraction is approximately 0.1 except at low aerosol concentrations. Verheggen et al.’s (2007) study is based on a very large dataset with over 900h of in-cloud measurements. In other observational studies, based on fewer data, this effect is less pronounced. Gillani et al. (1995) find higher scavenged fractions (with a median of 85-90%) for total aerosol number concentrations up to 600 cm$^{-3}$ in continental stratiform clouds, and a decrease only for concentrations above this value. Their values are based on measurements of unactivated accumulation mode particles (0.1 – 1μm in radius) and cloud droplets. In a previous campaign at the Jungfraujoch, no dependence of the activated fraction on the aerosol particle concentration was found (Baltensperger et al., 1998), and the average activated fraction for $r > 50\text{nm}$ from four events during a campaign in October/November 1993 was 0.48.

The dependency of $F_N$ on the total aerosol concentration has been analyzed for simulations CTL and AP in Figures 3.16 and 3.17. Only the definition $F_{\text{CTL},1}$ (3.9) for the scavenged fraction in simulation CTL is shown here, as $F_{\text{CTL},2}$ is not sensitive to the total particle number. In Figures 3.16(a) and (b) and 3.17(a) and (b), we distinguish between clouds containing liquid droplets and pure ice clouds. As the minimum cloud droplet number concentration in ECHAM5 is prescribed to 40 cm$^{-3}$, the scavenged fraction is constrained in the range of low aerosol concentrations. For simulation CTL, it is 1 if $N_{\text{ aer,total}}(r > 50\text{nm})$ is below 40 cm$^{-3}$, and is enveloped by a function of the form $1/x$ above this threshold. For simulation AP, values of $(N_1 + N_i + N_{\text{int}}(r > 50\text{nm}))$ below 40 cm$^{-3}$ are impossible in liquid/mixed-phase clouds. Above 40 cm$^{-3}$ the scavenged fraction for liquid/mixed clouds is enveloped by a function of the
Figure 3.15: Scavenged fraction $F_{AP}$ versus temperature for simulation AP. The boxplot (b) is based on the same data as (a). The boxes and whiskers indicate the 10th, 25th, 50th, 75th and 90th percentiles. The mean per temperature interval is marked with an asterisk. The number of data points per temperature interval is given above each box. Observations by Henning et al. (2004) and Verheggen et al. (2007) are included for comparison.

Figure 3.16: Scavenged fraction $F_{CTL,1}$ versus aerosol concentration for simulation CTL, for temperatures above ((a) and (c)) and below ((b) and (d)) −5°C. The dashed line gives the scavenged fraction for the minimum droplet concentration of 40 cm$^{-3}$. The boxplots (c) and (d) are based on the same data as the plots above, but are displayed only for $N_{vol}(r > 50\text{nm}) \leq 200 \text{ cm}^{-3}$. The boxes and whiskers indicate the 10th, 25th, 50th, 75th and 90th percentiles. The mean per concentration interval is marked with an asterisk. Observations by Verheggen et al. (2007) are included for comparison.
3.5 Comparison with observations

Figure 3.17: As Figure 3.16, but for simulation AP.

3.5.7 Sensitivity studies on cloud droplet activation

The comparison with marine boundary layer aerosol number concentrations (section 3.5.1) and marine stratocumulus droplet concentrations (section 3.5.5) has revealed the discrepancy that while the aerosol concentration is captured much better in simulation AP, the cloud droplet number concentrations are overestimated by more than a factor of 2. Furthermore, the cloud droplet burden in both simulations CTL and AP is significantly higher than retrieved from satellite observations by Han et al. (1998). The simulated global distribution of the cloud droplet burden deviates significantly from the observations (Figure 3.18). For this Figure, only values from cloudy gridpoints with liquid cloud tops are analyzed, same as in Han et al.’s (1998) retrieval, and the simulated grid-mean cloud droplet burden is divided by the total cloud fraction at each gridpoint. While the retrieval exhibits a clear land-sea contrast with lower values over the oceans, this is less pronounced in simulation CTL and even less in simulation AP. Furthermore, the simulations show local maxima of the cloud droplet burden in the middle of oceans (e.g. in the South Indian ocean), where the observations report minima. The droplet burdens in the stratocumulus regions off the South African and South American west coasts are most severely underpredicted.

Mis-predicted cloud droplet burden can either be due to errors in the in-cloud droplet concentration or in the cloud thickness. Here we analyze possible problems in the implementation of the droplet nucleation
Figure 3.18: Annual mean cloud droplet number burden, only for clouds with liquid cloud tops, from simulations CTL and AP in comparison to retrievals by Han et al. (1998). White: no data.

Figure 3.19: Zonal annual mean cloud droplet number burden from different simulations in comparison to retrievals by Han et al. (1998).
Figure 3.20: Droplet concentrations \( N_l \) versus concentration of soluble aerosol particles with a radius larger than 35 nm \( (N_{aer>35nm}) \). The continuous line is the 1:1 relation, and the dashed line gives the maximum activation \( N_{l,max} \) according to Lin and Leaitch (1997).

Parameterization. In simulation CTL, it is based on Lin and Leaitch (1997) (see also section 2.2.2).

\[
N_{l,new} = N_{l,old} + 0.1 \times \left( \frac{w (N_{aer>35nm} - N_{l,old})}{w + \alpha (N_{aer>35nm} - N_{l,old})} \right)^{1.27} \tag{3.12}
\]

\( N_{l,new} \) and \( N_{l,old} \) are the new and old cloud droplet number concentrations in \( \text{cm}^{-3} \), \( w \) is the updraft vertical velocity in \( \text{cm} \, s^{-1} \), \( \alpha \) is 0.023 \( \text{cm}^4 \, \text{s}^{-1} \), and \( N_{aer>35nm} \) the concentration of aerosol particles in the soluble modes with a radius larger than 35 nm. Activation is calculated if one or more of the following conditions are fulfilled: (a) \( N_{l,old} \) is below the minimum value of 40 \( \text{cm}^{-3} \), (b) the tendency of liquid water is positive (from condensation, vertical diffusion or transport), (c) the cloud cover is larger than in the previous timestep, or (d) the air is supersaturated with respect to water. Furthermore, if detrainment of liquid water from convective clouds occurs, additional activation is calculated with a lower cut-off radius of 25 nm for activation (Lohmann, 2007).

\[
N_{l,new}' = N_{l,new} + 0.1 \times \left( \frac{w (N_{aer>25nm} - N_{l,from \, 35nm})}{w + \alpha (N_{aer>25nm} - N_{l,from \, 35nm})} \right)^{1.27} \tag{3.13}
\]

Here \( N_{l,from \, 35nm} \) is the maximum possible droplet number from particles with dry radius \( r > 35 \) nm to be activated in the same timestep (this number is calculated even if the conditions for activation in stratiform clouds are not fulfilled). Equations (3.12) and (3.13) are calculated at cloud base and the droplet concentration is applied for the whole vertical cloud extent.

From Lin and Leaitch’s (1997) original formulation, a maximum droplet concentration can be calculated in the limit of high updraft velocities: \( N_{l,max} = 0.1 \times N_{aer>35nm}^{1.27} \). The above-described treatment of cloud droplet activation bears the risk of predicting values higher than \( N_{l,max} \). First, the vertical mixing assumption in clouds leads to droplet concentrations which are based on high aerosol concentrations at cloud base, although in higher levels less aerosols are available. Second, the lower cut-off diameter in convective clouds is an ad-hoc assumption and is not consistent with the measurements by Leaitch et al. (1996) on which the parameterization is based. Leaitch et al. (1996) did not determine the lowest activated particle size. Third, the formulation in simulation CTL can lead to repeated activation in subsequent timesteps, leading to an overestimation of cloud droplet concentrations. Figure 3.20(a) illustrates that the in-cloud droplet concentration is in many cases higher than \( N_{l,max} \), and in several cases even higher than \( N_{l,aer>35nm} \). This behavior seems unphysical.

In simulations CTL-nuc and AP-nuc, the additivity of the cloud droplet activation has been changed. Instead of adding the new activated particle number to the preexisting particles, the maximum of the old and new droplet number is chosen. For air detrained from convective clouds, the same cut-off radius of 35 nm is set. Activation is now calculated for each level separately instead of only at cloud base.

\[
N_{l,new} = \text{Max} \left( N_{l,old}, 0.1 \times \left( \frac{w (N_{aer>35nm} - N_{l,old})}{w + \alpha (N_{aer>35nm} - N_{l,old})} \right)^{1.27} \right) \tag{3.14}
\]
3 Aerosol Processing: GCM

\[
N_{l,new}'' = \text{Max} \left( N_{l,new}, 0.1 \times \left( \frac{w \left( N_{\text{crit} > 35 \text{nm}} - N_{l,new} \right)}{w + \alpha \left( N_{\text{crit} > 35 \text{nm}} - N_{l,new} \right)} \right)^{1.27} \right)
\]  \hspace{1cm} (3.15)

As seen in Figure 3.20(b), this reduces the cases of \( N_l > N_{\text{crit} > 35 \text{nm}} \), but still in many cases the droplet concentration lies above the maximum predicted from Lin and Leaitch’s (1997) formulation. This is due to higher aerosol concentrations in previous timesteps. With \( N_l \) as a prognostic variable, which is determined by the aerosol at the timestep of cloud formation and then evolves as a function of the cloud microphysical processes, but is not recalculated at each timestep for changing aerosol concentrations, such inconsistencies are hard to avoid. The global cloud droplet burden decreases by 25% (CTL-nuc) and 12% (AP-nuc), respectively (Table 3.2 and Figure 3.19). As a second test, the cut-off radius has been doubled in simulation AP-cut to 70 nm (also for detrainment from convective clouds), but with keeping the same additive treatment as in simulation AP. This had less of an effect on the droplet burden (a reduction of only 9% on global average) than changing the additivity and calculating nucleation in each level. Neither of these changes reduces the global mean in-cloud droplet burden down to Han et al.’s (1998) value of \( 4 \times 10^{12} \text{m}^{-2} \), nor do they significantly improve the global distribution. The fact that the cloud droplet burden is overestimated especially over the oceans, although Lin and Leaitch’s (1997) parameterization has been derived from measurements in marine stratus clouds, hints at more basic problems in the application of this formulation, or alternatively in the cloud dynamics and estimates of the cloud thicknesses. A different activation parameterization which takes into account the competition effect between activated particles of different sizes (Abdul-Razzak and Ghan, 2000) will be implemented in ECHAM5-HAM in future.

3.6 Conclusions

An explicit treatment of in-cloud aerosol particles in the global aerosol-climate model ECHAM5-HAM has allowed a global assessment of the turnover of aerosol particles in stratiform clouds. Compared to previous estimates (Pruppacher and Jaenicke, 1995), ECHAM5-HAM simulates a lower number of cycles through clouds (0.43 compared to 3). The discrepancy is due to the fact that aerosol processing in convective clouds is not yet included in ECHAM5-HAM, and to an overestimation of the global liquid water path by Pruppacher and Jaenicke (1995). Recalculating Pruppacher and Jaenicke’s (1995) estimate for stratiform clouds yields 0.4 cycles through clouds, which is in good agreement with ECHAM5-HAM.

The rates of aerosol mass transfer into cloud droplets and ice crystals can be analyzed, and it is found that nucleation scavenging dominates over collision scavenging for sulfate, sea salt and mineral dust, while the two processes are of similar importance for black carbon and organic carbon. Aqueous-phase formation of sulfate is the most important source of atmospheric sulfate particles. As in the simulation with explicit aerosol processing (AP) in general fewer particles are scavenged into the cloud phase than in the control simulation CTL, more particles are available for activation, resulting in higher cloud droplet concentrations. An enhancement of the autoconversion rate is necessary in order to achieve an equilibrated radiation balance, and this in turn reduces the liquid water path. Higher oxidation rates of \( \text{SO}_2 \) in the gas phase instead of in the aqueous phase entails higher nucleation rates of sulfate particles, and thus also higher particle concentrations in the small size range.

Comparison to different observations reveals several inconsistencies. While the marine boundary layer accumulation mode number concentrations are better reproduced in simulation AP than in simulation CTL, the opposite is true for the droplet concentrations in marine boundary layer clouds. Sea salt mass concentrations are well simulated over ocean, but overestimated over land. The wet and dry diameter of the accumulation mode is overestimated especially over ocean. Total volume burden and aerosol optical depth at a large number of AERONET stations is not well simulated. The scavenged particle number fraction at the Jungfraujoch is satisfactorily simulated in both simulation CTL and AP, with a general overestimation at low temperatures in CTL and an underestimation at warm temperatures in AP. While for CTL this is analyzed only diagnostically and the scavenged fraction for wet deposition is fixed to excessively high values at low temperatures, the scavenged fraction in AP is actually directly applied for wet deposition calculations. Finally, it is explored how the treatment of cloud droplet activation can be modified in order to simulate a more realistic global cloud droplet burden, but with the currently implemented parameterization by Lin and Leaitch (1997) no satisfactory results are achieved, although the in-cloud droplet concentrations in the control simulation agree well with satellite retrievals by Bennartz (2007).
3.6 Conclusions

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4 Modelling of Arctic mixed-phase clouds: M-PACE

4.1 Arctic clouds

The Arctic is particularly vulnerable to climate change, with an annual mean warming roughly twice as large as on global average (Christensen et al., 2007). However, predictions are particularly difficult because of the complex interactions between atmosphere, ocean, sea ice and permafrost land surfaces. The region is extensively covered in all seasons by clouds of different characteristics, with a maximum in summer, when low-level stratus clouds prevail in the Arctic basin. Interactions with short- and longwave radiation are different from other locations because of the low surface temperature and high albedo of underlying pack ice, or the strong surface forcing from relatively warm open ocean waters, and because of the weak daily and strong seasonal cycle of solar irradiance (Houze, 1993).

Tsay and Jayaweera (1984) have classified Arctic stratus clouds observed over the Beaufort Sea in 1980 into three categories, according to the meteorological conditions during their formation. They distinguish three types:

1. Clouds occurring in cold air outbreaks flowing over a warmer sea ice surface, formed by a convective-type process. This was observed in one out of seven cases.

2. Clouds forming in warm moist air from the south, flowing over the sea ice surface. They form very near the surface and can be multi-layered.

3. Clouds forming as a result of occluded and surviving frontal systems, with complex morphology.

Type 1 is similar to cases of stratus clouds forming when cold polar air flows south over the open ocean, which is warmer than the sea ice surface. Such a case has been observed and modelled by Müller et al. (1999). The input of sensible and latent heat from below leads to thermal instability and convection. Together with wind shear, this results in the formation of wind-parallel boundary layer rolls. Type 2 is the typical and very frequent Arctic summertime layered cloud, as also described by Curry et al. (1996) and Houze (1993).

In the last ten years, important field campaigns over the Beaufort Sea and the North Slope of Alaska helped advancing our understanding of Arctic stratus clouds: SHEBA (Surface Heat Budget of the Arctic Ocean (Uttal et al., 2002)) from October 1997 to October 1998 and FIRE-ACE (First ISCCP (International Satellite Cloud Climatology Project) Regional Experiment - Arctic Clouds Experiment (Curry et al., 2000)) from April to July 1998. Recently, more data were sampled during the Mixed-Phase Arctic Cloud Experiment (M-PACE), see below (section 4.2).

From SHEBA observations, Intrieri et al. (2002) concluded that clouds exert a net warming effect on the surface during most of the year, because the longwave fluxes dominate over the shortwave radiation except for a short period in summer. Arctic stratus clouds are susceptible to aerosol indirect effects because of the usually very low aerosol concentrations. In winter or spring, Arctic haze events with transport of pollutants, primarily from Eurasia, to the Arctic, lead to a strong increase in cloud condensation nuclei (Law and Stohl, 2007). In liquid clouds, this results in an increase of cloud droplets and of the shortwave cloud albedo (the first aerosol indirect effect). For thin liquid Arctic layer clouds, the increase in cloud droplets can also influence the longwave emissivity of the clouds (Lubin and Vogelmann, 2006; Garrett and Zhao, 2006). This effect is usually not considered for other regions, because for thicker liquid clouds, the emissivity is close to the blackbody value of 1, and aerosols have little effect on the cloud interactions with longwave radiation. Increased Arctic cloud longwave emissivity due to pollution leads to a surface warming in winter and early spring, but during late spring, the increase in cloud albedo and the related shortwave cooling exceeds this warming (Lubin and Vogelmann, 2007).

In mixed-phase and ice clouds, aerosol indirect effects are more complex. During SHEBA, mixed-phase clouds occurred on average 41% of the time, with a maximum in fall (>70%). All-ice clouds were observed 20% of the time, without a pronounced seasonal cycle, and all-liquid clouds 19% of the time,
most frequently in summer (Shupe et al., 2005). While self-maintaining mechanisms in liquid Arctic summer clouds have been identified early (Houze, 1993), the persistence of mixed-phase clouds is still not fully understood. Mixed-phase clouds are colloidal unstable, because the Wegener-Bergeron-Findeisen Process should lead to a steady increase of the ice fraction, precipitation and finally cloud dissipation (Harrington et al., 1999). Often clouds are heterogeneous on small spatial scales. Thin liquid layers at the cloud top which precipitate ice are frequently observed in the Arctic (Hobbs and Rangno, 1998).

Even in slightly (cloud top temperature of 0 to $-10^\circ$C) to moderately ($-10$ to $-20^\circ$C) supercooled clouds, high ice crystal concentrations (> 50 l$^{-1}$) are observed (Rangno and Hobbs, 2001). They can exceed the ice nuclei concentration parameterized by Meyers et al. (1992) by more than a factor of 10 and might be caused by ice splintering during riming or fragmentation of ice crystals and shattering of drops during freezing (Rangno and Hobbs, 2001). Harrington et al. (1999) showed that simulated stability of a mixed-phase cloud layer is strongly dependent on ice nuclei concentrations. Ice nuclei concentrations measured in-cloud for one case during FIRE-ACE (Curry et al., 2000) and averaged over the M-PACE campaign (Prenni et al., 2007) are lower than 1 l$^{-1}$. Therefore, depletion of ice nuclei by nucleation scavenging is a potentially important process in Arctic clouds (Morrison et al., 2005).

### 4.2 The Mixed-Phase Arctic Cloud Experiment (M-PACE)

From 27 September to 22 October 2004, an intensive observational period was conducted by the US Department of Energy Atmospheric Radiation Measurement (ARM) program at the North Slope of Alaska (Barrow): the Mixed-Phase Arctic Cloud Experiment (M-PACE). Verlinde et al. (2007) give an extensive overview over the aims, synoptic situations, equipment and first results. Mixed-phase clouds were chosen as the focus of this study, as substantial uncertainties prevail regarding the coupling between cloud microphysics, radiation and cloud dynamics.

The seasonal cycle of cloud cover at the North Slope of Alaska obtained from satellite and ground-based observations is shown in Figure 4.1. The cloud cover is lowest in winter (40-60%) and highest at the end of summer, in August and September (80-90%). A secondary maximum is found in spring (May). Values in June and July are lower than reported by the same authors for the whole Arctic region (defined as $60^\circ$N to $90^\circ$N) (Kato et al., 2006). In October 2004, the sea ice north of Alaska extended less far than the median ice edge since 1978 (Ketterer and Knowles, 2002). The Beaufort Sea was largely ice-free, especially at the beginning of October (Figure 4.2).

A model intercomparison study (Klein et al., 2007) for Single Column Models (SCM), Cloud Resolv-
4.3 Period A

During period A (October 5, 14 UTC to October 8, 14 UTC), a small midlevel trough brought considerable amounts of moisture to the north coast of Alaska (Figure 4.3). The resulting clouds over Barrow have a complex, multi-layered structure with thin liquid layers at the base of small temperature inversions and ice present throughout the cloud, probably sedimenting between the different liquid layers (Klein et al., 2007). In the typology by Tsay and Jayaweera (1984), this case falls into category 3.

Results from the model intercomparison for period A will be described by Morrison et al. (subm.). Initial conditions and large-scale forcings are provided by the ARM variational analysis (Xie et al., 2006). Three different experiments have been set up:

1. standard simulation (A1)
2. without ice (A2) - not shown here, because ECHAM5 simulates a similar cloud evolution as in simulation A1
3. enhanced vertical resolution: 98 instead of 31 vertical levels (A3)

Figure 4.4 shows ECHAM5 results compared to ground-based observations. In both simulations A1 and A3, ECHAM5 simulates a multi-layered cloud from approximately October 6, 00 UTC to October 7,
Figure 4.4: Model results for M-PACE period A. Observations are Active Remotely-Sensed Cloud Locations (ARSCL), microwave radiometer cloud liquid water path, and precipitation measured at Barrow (data provided together with the M-PACE forcing data (Klein et al., 2007)).

18 UTC. In the vertical, the cloud extends from 950 to 600 hPa, which is consistent with observations. The observed cloud is more compact than the simulated clouds, which consist of 2–3 (A1) and 3–5 (A3) distinct layers. Additionally, the model simulates fog in the lowest layer and an ice clouds at upper levels, which are not observed. In accordance with observations, the lower cloud partly dissipates around 15 UTC on October 7. In simulation A3, a thin layer persists until the cloud deepens again on the second half of October 8. This layer is missed in the lower-resolution simulation A1. The evolution of liquid water path (LWP), ice water path (IWP) and precipitation does not temporally correlate with observations, but the mean values agree reasonably for simulation A1. On average over the simulation period, the observed liquid water path is 64 g m\(^{-2}\), simulation A1 gives 85 gm\(^{-2}\), and simulation A3 is markedly higher with 146 g m\(^{-2}\). No observations of the ice water path are available. The model simulates low values (< 5 g m\(^{-2}\)) except during the occurrence of the upper-level cloud. Light precipitation is both observed (on average 0.05 mm h\(^{-1}\)) and simulated (0.07 (A1) and 0.09 (A3) mm h\(^{-1}\)).

4.4 Period B

During the M-PACE model intercomparison period B (October 9, 17 UTC to October 10, 5 UTC), a single-layer boundary layer cloud was observed. It formed as a result of a cold-air outbreak, similar to the first type of Arctic stratus described by Tsay and Jayaweera (1984) (see section 4.1). Cold near-surface air from the sea ice was advected over the warmer ocean for a distance of about 500 km before reaching the North Coast of Alaska. This north-easterly flow can be clearly seen on the Terra MODIS image from October 9 (Figure 4.5). As the air leaves the pack ice, it encounters an ocean surface which is several degrees warmer. Idealized profiles of potential temperature, liquid and total water are displayed in Figure 4.6. This substantial heating and moisture input from below triggers convection and mixing in the boundary layer, and liquid water condenses below the inversion. The clouds form very soon after the air reaches the open water, and subsequently become thicker with organized convection structures. First convection rolls as described by Müller et al. (1999) emerge, which later broaden and finally (west of Barrow) evolve into cellular convection structures. The idealized profiles, which are derived from a sounding at Barrow (Klein et al., 2007), show a dry adiabatic layer below the cloud, a moist adiabatic layer in the cloud and a very sharp inversion above. The cloud is roughly 600 m thick. These idealized profiles have been used to initialize the models. Furthermore, surface fluxes of sensible and latent heat
are prescribed. The models are set up over an ocean surface.

Results from the model intercomparison for period B will be described by Klein et al. (subm.). Here I show a few results from ECHAM5 simulations. As for period A, three different experiments have been set up:

1. standard simulation (B1)
2. without ice (B2)
3. enhanced vertical resolution: 98 instead of 31 vertical levels (B3)

ECHAM5 simulates a shallow boundary layer cloud, consisting almost exclusively of liquid water (Figure 4.7). In simulations B1 and B2, the liquid water content (LWC) remains close to the initial profile with a maximum value of 0.35 g m\(^{-3}\). In simulation B3, layers with higher liquid water content (0.5 g m\(^{-3}\)) form. During the second half of the simulation period, a thin, lower cloud layer (below 920 hPa) develops, which is not consistent with observations.

With a prescribed aerosol number concentration of 74 cm\(^{-3}\) particles in the accumulation and coarse modes, obtained from below-cloud particle counter measurements (Klein et al., 2007), the simulated cloud droplet number concentration \((N_l)\) is approximately 30 cm\(^{-3}\). This is a low value even for marine conditions. Quante (2004) gives 45–300 cm\(^{-3}\) as a typical range of hydrometeor concentrations in marine stratocumulus. Note that in ECHAM5, the cloud droplet number concentration is normally constrained to a minimum value of 40 cm\(^{-3}\), but for these simulations the minimum value has been reduced to 1 cm\(^{-3}\). Measured cloud droplet number concentrations at October 9, 20–22 UTC and October 10, 00–03 UTC are 26 and 72 cm\(^{-3}\), respectively, which large variability between individual spirals (McFarquhar et al., 2007). The simulated cloud drop effective radius \(r_{\text{eff},l}\) is 15–17 µm in the upper cloud layers, larger than the observed values which range from 7–11 µm.

While the liquid microphysical variables are roughly in accordance with observations, large discrepancies are found in the ice phase. The model is initialized with only liquid water; thus, ice has to be formed by heterogeneous nucleation. In ECHAM5, the ice water content (IWC) is underestimated by an order of magnitude, while ice crystal number concentrations \((N_i)\) are overestimated by an order of magnitude. As a result, the ice crystal effective radius \(r_{\text{eff},i}\) (which is parameterized as a function of the ice water content (Lohmann and Roeckner, 1996)), is only half of the observed value of 25 µm. The problem is of technical nature: as the ice diffuses rapidly between the vertical layers, the lower threshold of ice water mixing ratio for growth by deposition (0.5 mg kg\(^{-1}\)) (Lohmann et al., 2007), which is calculated before heterogeneous freezing, is never reached. Thus, ice mass is added only by heterogeneous freezing. The ratio IWC/LWC is of the same order of magnitude as \(N_i/N_l\). It is expected that this is only a problem of the single-column model. As ice microphysical processes are rather unimportant for the simulated cloud properties for this case, simulation B2 (without any ice formation) is very close to simulation B1.
Figure 4.6: Initial profiles of potential temperature $\theta$, liquid water mixing ratio $q_l$ and total (liquid+vapor) mixing ratio $q_t$ for period B. The diamond marks the ocean surface potential temperature.
Figure 4.7: Model results for M-PACE period B, averaged over the 12h simulation period. Simulations B1 and B2 lie close together. Observations are aircraft data from McFarquhar et al. (2007), averaged over several vertical spirals for two flights during period B. The standard deviations correspond to deviations of the vertically averaged value of each spiral to the average value over all spirals per flight.
Model difficulties in the liquid/ice partitioning for M-PACE period B have been reported previously, but authors have come to contradicting conclusions. On the one hand, Fridlind et al. (2007) find in Large-Eddy Simulations with the model DHARMA (Distributed Hydrodynamic Aerosol-Radiation-Microphysics Application), that the observed ice nuclei concentrations are insufficient by 3 orders of magnitude to explain the observed ice water content. They put forward two exotic mechanisms as possible explanation: ice nuclei which are emitted from evaporating droplets (evaporation nuclei), observed by Rosinski and Morgan (1991), or drop freezing during evaporation, which has never been reported before. On the other hand, Prenni et al. (2007) find good agreement between the liquid water path observations and RAMS (Regional Atmospheric Modeling System) simulations, if M-PACE ice nuclei measurements are used. With the standard ice nucleation formulation of RAMS, ice formation was vastly overestimated and nearly no liquid water remained after 12 hours of simulation.
5 Global influence of dust mineralogical composition on heterogeneous ice nucleation in mixed-phase clouds

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Abstract

Mineral dust is the dominant natural ice nucleating aerosol. Its ice nucleation efficiency depends on the mineralogical composition. We show the first sensitivity studies with a global climate model and a three-dimensional dust mineralogy. Results show that depending on the dust mineralogical composition, coating with soluble material from anthropogenic sources can lead to quasi-deactivation of natural dust ice nuclei. This effect counteracts the increased cloud glaciation by anthropogenic black carbon particles. The resulting aerosol indirect effect through the glaciation of mixed-phase clouds by black carbon particles is small (+0.1 Wm⁻² in the shortwave top-of-the-atmosphere radiation on the Northern Hemisphere).

5.1 Introduction

Aerosol particles interact with clouds in several ways. Their influence on liquid clouds has been studied extensively (e. g. Twomey, 1974; Albrecht, 1989). Anthropogenic aerosols add to the number of condensation nuclei available for droplet nucleation. An increased number of droplets at fixed cloud water content leads to a higher cloud albedo. Furthermore, the suppression of precipitation through more but smaller droplets increases the cloud lifetime. The magnitude of the different indirect aerosol forcings is still unclear (Lohmann and Feichter, 2005).

In mixed-phase clouds a third effect, the cloud glaciation effect, has been hypothesized (Lohmann, 2002a). Mixed-phase clouds, containing both liquid and frozen water, are frequent in mid-latitude and polar regions (Quante, 2004). At temperatures below 0°C, water droplets do not freeze spontaneously, but can persist as supercooled liquid until approximately −38°C, the homogeneous freezing temperature. At warmer temperatures, freezing of droplets occurs only through heterogeneous nucleation on ice nuclei. Ice nuclei are predominantly insoluble particles, such as mineral dust or metallic material (Richardson et al., 2007). The relative importance of the different nucleation modes (deposition, immersion, condensation and contact freezing) is still unclear. For immersion freezing an ice nucleus within a supercooled cloud droplet initiates the freezing process, while during contact freezing a supercooled droplet collides with a dry ice nucleus, such that the freezing process is initiated from the outside. Deposition refers to the direct growth of ice from the vapor on a dry ice nucleus, while for condensation freezing first a liquid layer condenses on a particle, which subsequently initiates the freezing. Although these ice nucleation processes are more efficient, the lower the temperature and the higher the supersaturation over ice, observed ice crystal number concentrations are highly variable for a given temperature and do not generally increase with lower temperatures (Gultepe et al., 2001).

As long as unfrozen droplets remain, the in-cloud air is saturated with respect to supercooled liquid water, and thus is supersaturated with respect to ice. This leads to rapid growth of the ice crystals and evaporation of the droplets (Bergeron-Findeisen process) (Findeisen, 1938). Therefore precipitation formation is more efficient in mixed-phase clouds than in warm clouds (Rogers and Yau, 1989). For anthropogenic ice nuclei, such as black carbon (soot), this is called ‘glaciation indirect effect’. Natural ice nuclei, which have not increased through human activity, do not add to the glaciation indirect effect.

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Lohmann and Diehl (2006) have presented sensitivity experiments with the ECHAM4 general circulation model. This was the first general circulation model study which took the efficiencies of ice nuclei of different chemical composition into account. However, for freezing on mineral dust, only the two extreme cases of (1) all dust consisting of kaolinite, a less efficient ice nucleus, or (2) all dust consisting of montmorillonite, a highly efficient ice nucleus (Pitter and Pruppacher, 1973), were considered. They showed that whether dust is assumed to be composed of kaolinite or montmorillonite had significant implications for the total indirect aerosol effect, with the decrease in top-of-the-atmosphere net radiation between preindustrial and present-day climate varying between 1 ± 0.3 W m⁻² and 2.1 ± 0.1 W m⁻².

Whether anthropogenic activity actually increases or decreases ice nuclei (IN) concentrations, is a matter of debate. As reviewed by Szyrmer and Zawadzki (1997), emissions from vegetation fires, rocket exhaust, and certain industries have been shown to be prolific IN sources. However, some studies have also reported a decrease of IN concentration due to human activity. Braham and Spyers-Duran (1974) found significant deactivation of natural IN, measured at a fixed temperature of −16.6°C, in passing over a big city. Borys (1989) reported that pollution-derived Arctic haze aerosol has a factor of 10 − 1000 lower fraction of ice nucleating particles, measured at −25°C and water saturation, than the Arctic aerosol during clean conditions. Based on these data, Girard et al. (2005) formulated an IN reduction factor, depending on sulfuric acid mass concentration. Deactivation of ice nuclei can be due to the coating of insoluble, highly active contact IN with soluble material. With a coating of e.g. sulfuric acid or nitric acid, these particles could act as immersion or condensation nuclei instead, but these processes require lower freezing temperatures. For example, Pitter and Pruppacher (1973) measured a median freezing temperature for kaolinite particles in the contact mode of −12°C and of −28°C in the immersion mode. Dust particles often get coated with sulfate, e. g. by cloud processing, as observed in the Eastern Mediterranean by Levin et al. (1996). Ansmann et al. (2005) put forward deactivation of dust ice nuclei by coating and mixing with hygroscopic material as an explanation for an observed non-glaciated cloud at −9 to −16°C in the presence of a layer of Saharan dust transported over Europe. Storelvmo et al. (2008) were the first to test this hypothesis in a global model, assuming that dust particles act as immersion nuclei rather than contact nuclei if enough sulfuric acid is present in a gridbox for a coating of one monolayer. This resulted in less efficient freezing in present-day climate than in preindustrial climate.

In this study, we use the coupled aerosol-climate model ECHAM5-HAM, taking into account the coating of dust and black carbon particles with sulfate. By introducing monthly mean mineralogical compositions, which vary regionally depending on the origin of the dust, we aim at constraining the model between the two extreme cases of pure kaolinite or pure montmorillonite dust. For this we use a database by Claquin et al. (1999), who have compiled the mineralogical composition of mineral dust for all global source areas. For the clay fraction, i. e. particles smaller than 3 µm in diameter, the kaolinite, smectite (montmorillonite), illite, calcite and quartz mass fractions are given for a variety of soils. Kaolinite, smectite and illite together make up more than 70% in mass of the clay fraction of all investigated soils, for most cases even more than 85%. Illite is on average more abundant than smectite, but its ice nucleating properties have rarely been investigated. Hoffer (1961) found a very similar behavior of illite and montmorillonite with respect to immersion freezing. Therefore we do not distinguish these two minerals in the present study.

5.2 Model Description

We use the coupled aerosol-climate model ECHAM5-HAM (Stier et al., 2005) with a microphysics module that includes prognostic equations for the cloud droplet number concentration (Lohmann et al., 1999) and ice crystal number concentration (Lohmann, 2002b; Lohmann et al., 2007). The treatment of heterogeneous freezing is described below. The model we use is a preliminary version of ECHAM5.302 with only 4 instead of 6 shortwave spectral bands and without the cirrus scheme by Kä rcher and Lohmann (2002) and Lohmann and Kä rcher (2002), which gives a slightly different aerosol indirect effect than in Lohmann et al. (2007). However, this study mainly aims at comparing the different sensitivity scenarios and not at giving robust forcing estimates. Furthermore, only heterogeneous ice nucleation in mixed-phase clouds is studied here.

5.2.1 Heterogeneous Freezing Parameterization

The parameterizations of heterogeneous freezing processes between 0° and −35°C are based on the parameterizations used in ECHAM4 as described in Lohmann and Diehl (2006), but have been modified for this study to make use of the aerosol fields of ECHAM5-HAM and the offline dust mineralogy.
5.2 Model Description

Table 5.1: Parameters $a_x$ and $b_x$ for contact freezing (Diehl et al., 2006) and parameter $c_x$ for immersion freezing (Diehl and Wurzler, 2004).

<table>
<thead>
<tr>
<th>$x$</th>
<th>$a_x$ in $K^{-1}$</th>
<th>$b_x$</th>
<th>$c_x$ in $m^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>BC</td>
<td>0.0614</td>
<td>0.5730</td>
<td>0.00291</td>
</tr>
<tr>
<td>KAO</td>
<td>0.1007</td>
<td>0.6935</td>
<td>0.0615</td>
</tr>
<tr>
<td>MON</td>
<td>0.1014</td>
<td>0.3277</td>
<td>32.3</td>
</tr>
</tbody>
</table>

Table 5.2: The aerosol modes and components in ECHAM5-HAM (SO4=sulphate, BC=black carbon, OC=organic carbon, SS=sea salt, DU=dust).

<table>
<thead>
<tr>
<th>internally mixed/soluble modes</th>
<th>externally mixed/insoluble modes</th>
</tr>
</thead>
<tbody>
<tr>
<td>NS: nucleation mode, soluble</td>
<td></td>
</tr>
<tr>
<td>(SO4)</td>
<td></td>
</tr>
<tr>
<td>KS: aitken mode, soluble</td>
<td>KI: aitken mode, insoluble</td>
</tr>
<tr>
<td>(SO4, BC, OC)</td>
<td>(BC, OC)</td>
</tr>
<tr>
<td>AS: accumulation mode, soluble</td>
<td>AI: accumulation mode, insoluble</td>
</tr>
<tr>
<td>(SO4, BC, OC, SS, DU)</td>
<td>(DU)</td>
</tr>
<tr>
<td>CS: coarse mode, soluble</td>
<td>CI: coarse mode, insoluble</td>
</tr>
<tr>
<td>(SO4, BC, OC, SS, DU)</td>
<td>(DU)</td>
</tr>
</tbody>
</table>

We account for contact and immersion/condensation freezing. Homogeneous freezing and deposition nucleation is thought to be negligible in mixed-phase clouds.

The freezing rate via contact nucleation (cnt) is based on Young (1974a) and Cotton et al. (1986). It takes into account contact through Brownian diffusion, depending on the size of the droplets and of the aerosol particles. In contrast to Lohmann and Diehl (2006), we do not prescribe the initial mass of an individual ice crystal but set this mass equal to the droplet’s mass before freezing. This reflects the assumption that the droplets do not break up during freezing.

$$Q_{frz,cnt} \equiv -\frac{dN_i}{dt}_{frz,cnt} = D_{ap} 4\pi r_l N_{cnt} N_i$$  \hspace{1cm} (5.1)

$N_i$ is the concentration of liquid cloud droplets, $D_{ap}$ is the size-dependent Brownian aerosol diffusivity, and $r_l$ is the volume-mean droplet radius. $N_{cnt}$ is the number of contact nuclei. It is parameterized (in deviation from the original formulation by Young (1974a)) as following, taking into account the ice nucleation efficiencies of different aerosols:

$$N_{cnt} = \sum_x \text{Max} (1, \text{Min} (0, (-a_x(T-273.15K) - b_x))) \times \frac{N_{cnt,x}}{N_{aer,insol}} (N_i + N_i)$$  \hspace{1cm} (5.2)

with \( x = BC \text{ (black carbon)}, \text{ KAO \text{ (kaolinite)}}, \text{ MON \text{ (montmorillonite)}} \)

$N_i$ is the ice crystal number concentration, $N_{aer,insol}$ the total insoluble aerosol number concentration and $N_{cnt,x}$ a number concentration of the maximum available contact ice nuclei of material $x$. $T$ is the temperature in K. The parameters $a_x$ and $b_x$ (see Table 5.1) are related to the material’s effectiveness to act as ice nuclei in the contact freezing mode and have been determined from a compilation of experimental data by Diehl et al. (2006). Equation (5.2) has been derived from the parameterization of the number of frozen droplets as a function of the total (frozen plus unfrozen) droplet number, which is equivalent to $N_i + N_i$, by Diehl et al. (2006). Note that $N_{cnt}$ does not directly transfer into ice crystal concentrations, because the collision rate (equation (5.1)) is the limiting process for contact nucleation.

In ECHAM5-HAM, aerosol particles are represented as an external mixture of internally mixed modes (see Table 5.2). Coating of insoluble particles is simulated online. Only uncoated particles are assumed to act as contact nuclei. Therefore $N_{cnt,x}$ is obtained by summing up the number concentrations of all insoluble modes which contain that component and weighting this number with the surface area.

$$N_{cnt,BC} = \left( \frac{m_{BC,KI}}{m_{BC,KI} + m_{OC,KI}} \right)^{2/3} N_{KI}$$  \hspace{1cm} (5.3)
5 Dust and the glaciation indirect effect

\[ N_{\text{cnt,KAO}} = (N_{\text{AI}} + N_{\text{CI}}) x_{\text{KAO}} \] (5.4)  
\[ N_{\text{cnt,MON}} = (N_{\text{AI}} + N_{\text{CI}}) x_{\text{MON}} \] (5.5)

\( m \) are mass mixing ratios. The indices of the variables in these equations refer to the different modes and aerosol components in ECHAM5-HAM and are listed in Table 5.2. \( x_{\text{KAO}} \) and \( x_{\text{MON}} \) are the mass fractions of the dust components kaolinite and montmorillonite, which are not calculated in ECHAM5-HAM, but are obtained from an offline transport model as described in section 5.2.2. Here we assume that the different dust mineralogical components are externally mixed. As discussed by Balkanski et al. (2007), both external and internal mixtures of different minerals can possibly be generated during dust emission.

Immersion freezing (imm) is parameterized following Diehl and Wurzler (2004):

\[ Q_{\text{frz,imm}} = -\frac{dN_i}{dt} \bigg|_{\text{frz,imm}} = N_{a,\text{imm}} \exp(273.15 - T) \frac{\partial T}{\partial t} \frac{\rho_{\text{air}} q_l}{\rho_l} \] (5.6)

for \( \frac{\partial T}{\partial t} < 0 \)

Here \( \rho_{\text{air}} \) is the air density, \( q_l \) the liquid water density, \( q_l \) the cloud water mixing ratio, and the temperature \( T \) is given in Kelvin. The number of immersion nuclei, \( N_{a,\text{imm}} \), is related to a measured effectiveness \( c_x \) (see Table 5.1), again weighted by the number of potential ice nuclei of the respective material \( x \) (equation (5.7)). For immersion freezing only black carbon, montmorillonite and kaolinite in the soluble modes is considered because immersion freezing requires uptake of the particles into cloud droplets. We normalize the number of immersed black carbon, kaolinite and montmorillonite particles, \( N_{\text{imm},x} \), by the total number of particles larger than 35 nm in radius, which is the threshold for particles that can possibly be activated when using the activation parameterization by Lin and Leaitch (1997) in ECHAM5-HAM (Lohmann et al., 2007).

\[ N_{a,\text{imm}} = \sum_x c_x N_{\text{imm},x} \frac{N_{\text{aer,tot}}}{N_{\text{aer,tot}}} \] (5.7)

with \( x = \text{BC, KAO, MON} \)

The contributions \( N_{\text{imm},x} \) of the three different potential immersion freezing nuclei are calculated by:

\[ N_{\text{imm,BC}} = \left( \frac{m_{\text{BC,KS}}}{\rho_{\text{BC}}} \right)^{2/3} N_{\text{KS}} + \left( \frac{m_{\text{BC,AS}}}{\rho_{\text{BC}}} \right)^{2/3} N_{\text{AS}} + \left( \frac{m_{\text{BC,CS}}}{\rho_{\text{BC}}} \right)^{2/3} N_{\text{CS}} \] (5.8)

\[ N_{\text{imm,KAO}} = \left[ \frac{m_{\text{DU,KS}}}{\rho_{\text{DU}}} \right]^{2/3} N_{\text{AS}} + \left[ \frac{m_{\text{DU,AS}}}{\rho_{\text{DU}}} \right]^{2/3} N_{\text{AS}} + \left[ \frac{m_{\text{DU,CS}}}{\rho_{\text{DU}}} \right]^{2/3} N_{\text{CS}} \] x_{\text{KAO}} \] (5.9)

\[ N_{\text{imm,MON}} = \left[ \frac{m_{\text{DU,KS}}}{\rho_{\text{DU}}} \right]^{2/3} N_{\text{AS}} + \left[ \frac{m_{\text{DU,AS}}}{\rho_{\text{DU}}} \right]^{2/3} N_{\text{AS}} + \left[ \frac{m_{\text{DU,CS}}}{\rho_{\text{DU}}} \right]^{2/3} N_{\text{CS}} \] x_{\text{MON}} \] (5.10)

In the previous equations, \( V_{\text{KS}} \), \( V_{\text{AS}} \) and \( V_{\text{CS}} \) are the total dry volumes per kg air of aerosol particles in the Aitken, accumulation and coarse soluble/mixed modes, respectively.

\[ V_{\text{KS}} = m_{\text{SO4,KS}}/\rho_{\text{SO4}} + m_{\text{BC,KS}}/\rho_{\text{BC}} + m_{\text{OC,KS}}/\rho_{\text{OC}} \] (5.11)

\[ V_{\text{AS}} = m_{\text{SO4,AS}}/\rho_{\text{SO4}} + m_{\text{BC,AS}}/\rho_{\text{BC}} + m_{\text{OC,AS}}/\rho_{\text{OC}} + m_{\text{SS,AS}}/\rho_{\text{SS}} + m_{\text{DU,AS}}/\rho_{\text{DU}} \] (5.12)

\[ V_{\text{CS}} = m_{\text{SO4,CS}}/\rho_{\text{SO4}} + m_{\text{BC,CS}}/\rho_{\text{BC}} + m_{\text{OC,CS}}/\rho_{\text{OC}} + m_{\text{SS,CS}}/\rho_{\text{SS}} + m_{\text{DU,CS}}/\rho_{\text{DU}} \] (5.13)

The densities of the components are \( \rho_{\text{SO4}} = 1841 \text{ kg m}^{-3}, \rho_{\text{BC}} = 2000 \text{ kg m}^{-3}, \rho_{\text{OC}} = 2000 \text{ kg m}^{-3}, \rho_{\text{SS}} = 2165 \text{ kg m}^{-3} \) and \( \rho_{\text{DU}} = 2650 \text{ kg m}^{-3} \).

The cooling rate \( \frac{dT}{dt} \) in equation (5.6) is derived from the vertical velocity \( w \), which is obtained for liquid and mixed-phase clouds from \( w = \bar{w} + 1.33 \sqrt{\text{TKE}} \), with the large-scale vertical velocity \( \bar{w} \) plus
5.2 Model Description

Figure 5.1: Freezing rates as a function of temperature for $N_l = 100 \text{ cm}^{-3}$, $q_l = 0.03 \text{ g m}^{-3}$, $r_l = 4 \mu\text{m}$ and $dT/dt = -3.5 \cdot 10^{-4} \text{ K s}^{-1}$. Each of the curves assumes a different aerosol population, consisting purely of coated black carbon, uncoated black carbon, coated dust or uncoated dust particles, respectively. The fractions of maximum available ice nuclei over the total insoluble or total activated number of particles in equations (5.2) and (5.7) are therefore 1. The radius of the particles is 15 nm for black carbon and 370 nm for montmorillonite and kaolinite.

Figure 5.1 shows $Q_{\text{frz, cnt}}$ and $Q_{\text{frz, imm}}$ for pure black carbon, montmorillonite and kaolinite aerosol, for typical values of cloud properties and aerosol radii of freshly emitted particles from Stier et al. (2005). Contact freezing on montmorillonite sets in at highest temperatures ($-4^\circ\text{C}$), followed by kaolinite. Contact freezing on black carbon sets in below $-9^\circ\text{C}$, but then attains 2 orders of magnitude higher values than contact freezing on mineral dust. This is due to the higher aerosol diffusivity $D_{ap}$ of the Aitken mode black carbon particles compared to the accumulation mode dust particles.

Immersion freezing on montmorillonite becomes significant compared to contact freezing below $-25^\circ\text{C}$, immersion freezing on kaolinite only below $-31^\circ\text{C}$. Immersion freezing on black carbon is negligible for this set of cloud parameters until close to the homogeneous freezing threshold, where heterogeneous freezing is switched off in the model.

Freezing by Aitken-mode particles has not been definitely proven to occur in the atmosphere. On one hand, residuals from ice particles are often found to be on average larger than ambient particles (e.g., by Richardson et al. (2007)). On the other hand, Seifert et al. (2003) reported that small ($d < 100 \text{ nm}$) aerosol particles dominated the number density of residuals for measurements in cirrus clouds during the INCA campaign (Interhemispheric differences in cirrus properties from anthropogenic emissions), and related this finding to heterogeneous nucleation. Most laboratory studies investigate freezing only on ice nuclei with a diameter of 100 nm or larger. Sax and Goldsmith (1972), however, showed that silver iodide particles with a radius of 10 nm can act as efficient contact ice nuclei, colliding with droplets in free fall by Brownian diffusion.

The Bergeron-Findeisen process is parameterized as a threshold process (Lohmann et al., 2007). Once heterogeneous freezing leads to an ice water mixing ratio larger than the threshold value of 0.5 mg kg$^{-1}$, the remaining cloud droplets evaporate within one model timestep and the water mass is deposited onto the existing ice crystals. Ice crystal multiplication at temperatures between $-3$ and $-8^\circ\text{C}$ via the Hallett-Mossop process is also taken into account (Lohmann, 2002b).

### 5.2.2 Dust Mineralogical Composition

The dust mineralogy fields have been calculated offline with the chemistry transport model TM3 (Tegen et al., 2002). The mineral dust source strength is governed by the wind speed and soil parameters. The a subgrid-scale contribution estimated from the turbulent kinetic energy TKE (Lohmann et al., 2007). The turbulent contribution is usually much larger than the large-scale vertical velocity. For an adiabatic temperature gradient, this converts into the following temperature tendency (using the specific heat of air at constant pressure $c_p$ and the acceleration of gravity $g$):

$$
\frac{\partial T}{\partial t} = -\frac{g}{c_p} w
$$

(5.14)

This tendency is significantly larger than the large-scale temperature tendency $\partial \bar{T}/\partial t$ alone, which was used by Lohmann and Diehl (2006) for immersion freezing in ECHAM4. This increases the relative importance of immersion freezing compared to contact freezing.

Figure 5.1 shows $Q_{\text{frz, cnt}}$ and $Q_{\text{frz, imm}}$ for pure black carbon, montmorillonite and kaolinite aerosol, for typical values of cloud properties and aerosol radii of freshly emitted particles from Stier et al. (2005). Contact freezing on montmorillonite sets in at highest temperatures ($-4^\circ\text{C}$), followed by kaolinite. Contact freezing on black carbon sets in below $-9^\circ\text{C}$, but then attains 2 orders of magnitude higher values than contact freezing on mineral dust. This is due to the higher aerosol diffusivity $D_{ap}$ of the Aitken mode black carbon particles compared to the accumulation mode dust particles.

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source scheme of this model includes preferential sources of dust emission (topographic depressions), a seasonally changing vegetation masking dust emissions (Tegen et al., 2002) and satellite retrieved roughness lengths (Prigent et al., 2005). The computation of the mineralogical composition is based on Claquin et al. (1999), but is simplified by using only 27 major soil types, averaging the results by Claquin et al. (1999) for 107 soil types with area-weighted means. The mineralogical composition of the soil obtained in this way is shown in Figure 5.2 for the four components illite, smectite, kaolinite and calcite/quartz in the clay fraction. The clay fraction is defined here as fine particles up to a radius of 1.5 µm, even though this fraction also contains some quartz/calcite. This corresponds to the dust calculated by ECHAM5-HAM, which does not contain the large dust fraction.

The four components are transported as tracers in the model such that the mineralogical composition of airborne dust at each grid point can be calculated. TM3 was driven with meteorological fields from the ERA15 reanalysis for the year 1987. The timestep for computing the dust sources was 6 hours. The source fields were computed with 1.125° × 1.125° horizontal resolution.

ECHAM5-HAM also includes the Tegen et al. (2002) dust emission scheme. The mass flux of emitted dust is distributed into two lognormal modes with mass-median radii of 0.37 and 1.75 µm (Stier et al., 2005). These size distribution parameters have been obtained by a fit of multi-annual global mean emitted size distribution, represented by 24 size bins in the emission scheme by Tegen et al. (2002), with 3 lognormal modes. The super-coarse mode is neglected in ECHAM5-HAM because of its short lifetime. The dust emissions are attributed to the insoluble accumulation and coarse modes AI and CI (Table 5.2). Subsequent coating or coagulation with soluble material transfers the dust particles into the internally mixed modes AS and CS.

As in this study montmorillonite is also used as a proxy for illite (with respect to its freezing behavior), the montmorillonite fraction $x_{MON}$ (equations (5.5) and (5.10)) is defined as the ratio of the sum of the illite and smectite (which is a synonym for the montmorillonite clay group) mixing ratio divided by total clay mixing ratio. The kaolinite fraction $x_{KAO}$ (equations (5.4) and (5.9)) is the kaolinite mixing ratio divided by total clay mixing ratio.

The montmorillonite and kaolinite fractions were stored as monthly mean three-dimensional fields and used as input to ECHAM5-HAM. Figure 5.3 shows the annual mean kaolinite and montmorillonite fractions of the total atmospheric clay burden. Montmorillonite (49–60%) is more abundant than kaolinite (30–42%). In general, the kaolinite fraction is larger in the Southern Hemisphere because the kaolinite fraction in the soil is also largest in southern Africa, South America and Australia (see Figure 5.2). The montmorillonite fraction on average behaves reciprocally to kaolinite. The monthly fields on individual levels (not shown) exhibit a stronger variance.
5.2 Model Description

5.2.3 Evaluation of the Dust Mineralogical Composition Fields

The simulated mineralogical composition on the lowest level has been compared to various measurements of airborne particles and wet deposition samples. Figure 5.4 shows that overall the model simulates excessively uniform values for the kaolinite and montmorillonite fraction. Kaolinite tends to be overestimated, while montmorillonite (compared to the sum over illite and smectite measurements) shows less of a bias. This suggests that the calcite/quartz content of the measurements was higher than simulated. Partly this bias can be due to inclusion of particles larger than 3 μm in diameter, which tend to be richer in quartz, in the analysis. The observations refer to individual events at precise locations, while the model simulates the mean mineralogical composition for a larger area, averaged over a time period of one month. Additionally, the observations rely on a number of different techniques, so that individual values might not be comparable. The observations do not show any clustering regarding their large-scale source area (colour-coded in Figure 5.4). Therefore some lack of variability in the model is not surprising. In the coherent dataset by Stuut et al. (2005), which has been measured on a 8000 km cruise along the West African coast, a North-to-South increase of the kaolinite fraction has been observed. This is also reproduced in the model, although with an offset to higher values (see red squares in Figure 5.4, left plot).

5.2.4 Simulations

Three pairs of simulations have been carried out (see Table 5.3). In the simulation MIX the dust mineralogy described above has been used. In the sensitivity experiments KAO and MON, dust is assumed to be composed completely of one mineralogical component. Simulations with this assumption have also been studied by Lohmann and Diehl (2006). The simulations are run for 5 years (after a 3-months spin-up) in T42L19 resolution (2.8° × 2.8°, 19 vertical levels with a top at 10hPa, and a timestep of 15 minutes), using climatological sea surface temperatures and sea ice extent. Each simulation has been carried out once with present-day aerosol emissions and once with emissions representative for the preindustrial era, as described in Lohmann et al. (2007). For the preindustrial (PI) simulations, the greenhouse gas concentrations and sea surface temperatures are held constant at their present-day values in order to isolate the aerosol effect. Mineral dust aerosol is considered as natural aerosol (i.e., no changes in soil properties between preindustrial and present-day conditions are taken into account), although the percentage of mineral dust emission caused directly or indirectly by human activity is still under discussion. Tegen et al. (2004) estimate it at less than 10%. In our simulations, differences in the mineral dust emissions are solely caused by changes in the wind field.
Figure 5.4: Comparison of simulated and observed mineralogical composition at different locations. Colours refer to the presumed source areas of the measured samples (red: Africa, blue: East Asia, black: others).

Table 5.3: Sensitivity simulations.

<table>
<thead>
<tr>
<th>Simulation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>KAO</td>
<td>$x_{\text{KAO}} = 1$, $x_{\text{MON}} = 0$</td>
</tr>
<tr>
<td>MON</td>
<td>$x_{\text{KAO}} = 0$, $x_{\text{MON}} = 1$</td>
</tr>
<tr>
<td>MIX</td>
<td>$x_{\text{KAO}}$ and $x_{\text{MON}}$ read from input fields</td>
</tr>
<tr>
<td>MIX-nBC</td>
<td>$x_{\text{KAO}}$ and $x_{\text{MON}}$ read from input fields, $N_{\text{ext,BC}} = 0$ and $N_{\text{imm,BC}} = 0$</td>
</tr>
</tbody>
</table>
5.3 Results

5.3.1 Simulations of present-day climate

A thorough evaluation of ECHAM5-HAM is presented by Lohmann et al. (2007). Here we focus on differences between the three simulations MIX, KAO and MON. Annual zonal means of cloud liquid and ice water path, which can be compared to satellite observations, are shown in Figure 5.5. The three sensitivity studies differ only slightly. Figure 5.5(a) shows the liquid water path (LWP) over ocean surface compared to SSM/I retrievals by Greenwald et al. (1993), Weng and Grody (1994) and Wentz (1997). The global mean values amount to 67.6 g m$^{-2}$ in simulation MIX, 67.9 g m$^{-2}$ in simulation KAO and 67.4 g m$^{-2}$ in simulation MON. This falls within the observed range of 50 to 84 g m$^{-2}$. The zonal distribution is only partly captured in the simulations. Around the equator, ECHAM5-HAM simulates a too low liquid water path, while it is comparable to observations in the subtropics and midlatitudes.

The ice water path (IWP) is 27.8 g m$^{-2}$ in simulations KAO and MIX and 27.9 g m$^{-2}$ in simulation MON, which compares well to an estimate derived from ISCCP data by Storelvmo et al. (2008) of 29.5 g m$^{-2}$. The zonal distribution is well captured between 60°S and 60°N. At high latitudes, the comparison is more uncertain, because fewer satellite observations are available during the polar night.

In order to study the differences between the three sensitivity studies KAO, MON and MIX, we analyse the vertically integrated heterogeneous freezing rate, which reflects the effectiveness of freezing of the mineral dust. The zonal mean freezing rate, given as the number of frozen droplets in a vertical column per unit time and area, is highest in simulation MON and lowest in simulation KAO (Figure 5.6).

Figure 5.7 shows the global distributions of the vertically integrated number freezing rates. Here...
Figure 5.7: Vertically integrated total heterogeneous freezing rates on black carbon (BC) and dust (sum of immersion and contact freezing) from simulations MIX, KAO and MON.
5.3 Results

‘freezing on dust’ includes both contact and immersion freezing on dust, and ‘freezing on BC’ both contact and immersion freezing for black carbon. The freezing rates are in general larger at higher latitudes and larger over the continents than over the ocean, due to higher cloud droplet number concentrations over continents, resulting in more contact freezing. Additionally, the chance of cloud supercooling is enhanced at higher latitudes and also higher altitudes over orography. Overall, freezing by black carbon is simulated more frequently because of the high number concentrations and small sizes, thus high contact efficiencies, of insoluble black carbon particles. In some regions, e.g. South-East Asia, black carbon clearly dominates over freezing on dust. On the other hand, dust dominates the freezing at higher latitudes. Freezing on black carbon is very similar in the three simulations, such that the total freezing rate is essentially modulated by the freezing efficiency of dust.

Dust and BC particle number burdens (Figure 5.8), calculated by equations (5.3–5.5) and (5.8–5.10), are 1–2 orders of magnitude higher in the coated modes (which constitute the maximum available immersion nuclei) than in the uncoated modes (the maximum available contact nuclei). Black carbon is more abundant in number, because the particles are smaller. While no global observations of the fraction of coated particles are available, field campaigns at selected locations support these model results. E. g., Andreae et al. (1986) report that over remote oceans, 80–90% of the sampled silicate particles were associated with sea salt. Additionally, dust particles which are not internally mixed with sea salt, can react with volatile and semi-volatile atmospheric compounds. Asian dust particles collected over the sea of Japan during ACE-Asia, filtered to exclude particles associated with sea salt, were mixed with nitrate (on average 26% of the individually sampled particles), sulfate (18%) and chloride (18%), with only few particles containing more than one secondary acid (Sullivan et al., 2007). For black carbon, rapid aging is observed, too. At the high-altitude Alpine research station Jungfraujoch, with no local sources nearby, BC was found to be scavenged into hydrometeors to the same extent as the bulk aerosol, which suggests that BC was covered with soluble material (Cozic et al., 2007).

5.3.2 Anthropogenic Climate Forcing

A set of simulations for the preindustrial era has been run in order to study the sensitivity of the anthropogenic climate forcing by aerosols to heterogeneous freezing processes. The differences between present-day and preindustrial simulations are summarized in Table 5.4 and shown as zonal averages in Figure 5.9. They include both the direct aerosol effect and aerosol indirect effects on warm and mixed-phase clouds. Aerosol optical depth (AOD) increases from preindustrial to present-day times because of anthropogenic aerosol and precursor emissions. Cloud cover increases slightly because of the indirect cloud lifetime effect: More aerosol particles result in more cloud droplets, which (at constant liquid water content) have smaller sizes and therefore form precipitation less efficiently. Cloud lifetime increases and with it the mean cloud cover. Liquid water path increases because of the longer cloud lifetime. Ice water path remains nearly constant, because glaciated clouds dissipate quickly. The smaller cloud droplets also lead to increased scattering of incoming solar radiation (cloud albedo effect). The top-of-the-atmosphere (TOA) net radiation decreases because more radiation is scattered back to space. The global mean change in TOA net radiation is $-1.8\ W\ m^{-2}$ in simulation MON, $-2.0\ W\ m^{-2}$ in simulation MIX, and
Table 5.4: Global annual mean difference in liquid water path (LWP), ice water path (IWP), cloud droplet number burden ($N_{D}^{b}$), ice crystal number burden ($N_{C}^{b}$), cloud cover ($CC$) and top-of-the-atmosphere shortwave ($F_{SW}$), longwave ($F_{LW}$) and net ($F_{net}$) radiative fluxes between present-day and preindustrial simulations, with interannual standard deviations. $F_{SW}$ and $F_{net}$ are upward positive, $F_{LW}$ is downward positive.

<table>
<thead>
<tr>
<th>Simulation</th>
<th>KAO</th>
<th>MON</th>
<th>MIX</th>
<th>MIX-nBC</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta$LWP [g m$^{-2}$]</td>
<td>7.26 ± 1.00</td>
<td>6.72 ± 0.81</td>
<td>7.25 ± 0.89</td>
<td>7.54 ± 0.92</td>
</tr>
<tr>
<td>$\Delta$IWP [g m$^{-2}$]</td>
<td>0.23 ± 0.28</td>
<td>0.28 ± 0.29</td>
<td>0.20 ± 0.15</td>
<td>0.39 ± 0.18</td>
</tr>
<tr>
<td>$\Delta N_{D}^{b}$ [10$^{16}$ m$^{-2}$]</td>
<td>1.030 ± 0.066</td>
<td>1.037 ± 0.067</td>
<td>1.069 ± 0.045</td>
<td>1.117 ± 0.076</td>
</tr>
<tr>
<td>$\Delta N_{C}^{b}$ [10$^{13}$ m$^{-2}$]</td>
<td>0.058 ± 0.012</td>
<td>0.063 ± 0.007</td>
<td>0.064 ± 0.009</td>
<td>0.072 ± 0.009</td>
</tr>
<tr>
<td>$\Delta CC$ [%]</td>
<td>0.63 ± 0.30</td>
<td>0.48 ± 0.20</td>
<td>0.48 ± 0.24</td>
<td>0.49 ± 0.27</td>
</tr>
<tr>
<td>$\Delta F_{SW}$ [W m$^{-2}$]</td>
<td>2.24 ± 0.51</td>
<td>2.03 ± 0.41</td>
<td>2.25 ± 0.50</td>
<td>2.38 ± 0.43</td>
</tr>
<tr>
<td>$\Delta F_{LW}$ [W m$^{-2}$]</td>
<td>0.16 ± 0.31</td>
<td>0.22 ± 0.22</td>
<td>0.20 ± 0.18</td>
<td>0.43 ± 0.15</td>
</tr>
<tr>
<td>$\Delta F_{net}$ [W m$^{-2}$]</td>
<td>2.08 ± 0.43</td>
<td>1.80 ± 0.36</td>
<td>2.04 ± 0.56</td>
<td>1.86 ± 0.37</td>
</tr>
</tbody>
</table>

$-2.1$ W m$^{-2}$ in simulation KAO.

The differences between the three simulations are small and do not reflect a larger glaciation indirect effect in simulation KAO than in simulation MON. In the modeling study with ECHAM4 by Lohmann and Diehl (2006), assuming dust as a less efficient ice nucleus (kaolinite) increased the relative importance of freezing on black carbon and therefore increased the aerosol indirect effect on mixed-phase clouds and reduced the total aerosol indirect effect on the shortwave radiation. Only because of larger compensating emission of longwave radiation the net radiation was also most negative in the ECHAM4 simulation KAO.

Splitting up the global mean vertically integrated freezing rate into contributions by contact freezing on dust, immersion freezing on dust, contact freezing on black carbon and immersion freezing on black carbon (Figure 5.10) gives an explanation for this finding in the present study. Present-day total freezing rates are a factor of 4–6 higher than preindustrial freezing rates, partly because of higher droplet concentrations stemming from the aerosol indirect effect on warm clouds (especially over the continents) and partly because of higher black carbon concentrations. Immersion freezing on black carbon is negligible, but contact freezing on black carbon constitutes the major contribution to the total freezing rate. Its magnitude is very similar in KAO, MON and MIX. Uncoated black carbon particles, although they are far less abundant than coated black carbon particles (Figure 5.8), trigger most of the freezing events in present-day conditions. Large differences are seen in the freezing on dust, where the lower ice nucleation efficiency of kaolinite is visible. In simulations MON and MIX, contact freezing amounts to $\approx27\%$ of the freezing on dust in preindustrial climate and 15% in present-day climate. In contrast, in simulation KAO, it amounts to 80% (preindustrial) and 68% (present-day), although the absolute value is lower. Note that the number freezing rates in Figure 5.10 are not proportional to the mass of frozen water, as droplets in polluted regions (where freezing by black carbon is most important) are smaller than in remote regions and the frozen water mass is thus relatively smaller.

The partitioning between contact and immersion freezing on mineral dust is related to the aging of the particles. Only uncoated mineral dust particles are potential contact nuclei, while coated (aged) particles can potentially initiate immersion freezing. Figure 5.11 shows the simulated present-day and preindustrial dust mass burden and the contributions of insoluble (uncoated) and soluble/mixed (coated) modes. The total dust burden is relatively constant with 9.3 Tg (preindustrial) and 8.9 Tg (present-day), but the simulated contribution of uncoated dust to the total dust burden has shifted from 61% in preindustrial conditions to 48% in present-day conditions. Anthropogenic emissions of sulfate and SO$_2$ lead to a faster aging of the dust aerosol and thus a slightly larger removal rate. This shift is more pronounced on the Northern Hemisphere. Uncoated dust particles are found closer to the source regions and are on average larger than the aged, coated dust particle population, from which the largest particles have been removed by sedimentation and efficient wet deposition. Therefore the uncoated particles are less abundant in number (Figure 5.8).

Translated to dust ice nuclei concentrations, in present-day climate less potential contact nuclei and more potential immersion nuclei are available. As kaolinite ice nuclei are very inefficient in the immersion mode, this shift signifies a quasi-deactivation. Montmorillonite particles, on the other hand, are still active ice nuclei even when coated. Thus, the deactivation of kaolinite ice nuclei through coating by anthropogenic sulfate is the main difference between simulations KAO and MON. This deactivation dominates over the glaciation indirect effect (caused by anthropogenically increased black carbon concentrations) in simulation KAO. In simulation MIX, the immersion freezing stems from the contribution of the montmorillonite particles, and only the kaolinite part of the dust ice nuclei is quasi-deactivated.
5.3 Results

Figure 5.9: Difference between present-day and preindustrial runs for the zonal annual means of (a) aerosol optical depth (AOD), (b) cloud cover, (c) liquid water path (LWP) and (d) top-of-the-atmosphere (TOA) short wave (SW) radiation for the three experiments KAO, MON and MIX.

Figure 5.10: Vertically integrated global and annual mean heterogeneous freezing rates.
This effect was not taken into account by Lohmann and Diehl (2006).

### 5.3.3 Sensitivity to freezing on black carbon

As another sensitivity experiment, simulation MIX has been rerun (for present-day and preindustrial emissions) with suppression of both contact and immersion freezing on black carbon (simulation MIX-nBC). This can give an upper estimate of the glaciation indirect effect. Suppression of then freezing on anthropogenic black carbon only is not possible within the framework of ECHAM5-HAM, because this would require separate tracers for natural and anthropogenic black carbon. Results from the 5 year simulation are included in Table 5.4. However, contrary to expectations, the simulation MIX-nBC does not yield a larger change in the global mean liquid water path nor in the TOA shortwave radiation. In the zonal distribution, systematic differences between the Northern and the Southern Hemisphere are evident (Figure 5.12 and Table 5.5). In the Northern Hemisphere, if freezing on black carbon is suppressed (simulation MIX-nBC), the liquid water path, cloud cover and cloud droplet number burden increase more than in simulation MIX. Therefore the shortwave radiative forcing is stronger (more negative). When freezing on black carbon is included, more clouds glaciate and therefore the increase in liquid water path is lower. Note that the shift of dust ice nuclei from the contact to the immersion mode by coating with sulfate is taken into account in both simulations MIX and MIX-nBC. In the Southern Hemisphere, the heterogeneous freezing rates are in general much lower (see Figure 5.6) because larger areas are ocean. Changes in liquid water path, cloud cover, cloud droplet number burden and shortwave radiative forcing in simulations MIX and MIX-nBC show an opposite behavior than in the Northern Hemisphere. A detailed analysis has shown that this is not related to mixed-phase cloud processes, but to different wind-dependent emission fluxes of sea salt aerosol. Through dynamic, nonlinear feedbacks, surface winds decrease in simulation MIX-nBC, leading to less sea salt particles, while they increase in simulation MIX (Figure 5.13). This results in different concentrations of cloud condensation nuclei and slightly different effects on warm clouds, which are not the focus of this study. The decrease of surface winds is not a robust feature of model studies of the preindustrial era, and we therefore omit the Southern Hemisphere from further analysis of this simulation.

From this sensitivity study, we can give an upper estimate of the glaciation indirect effect for the Northern Hemisphere, where mixed-phase cloud processes are more important. The difference in shortwave radiative forcing between simulations MIX and MIX-nBC is +0.11 Wm$^{-2}$, which is offset by the difference in longwave radiation (less outgoing terrestrial radiation because of a higher cloud cover) of $-0.22$ W m$^{-2}$. The actual glaciation indirect is expected to be even slightly smaller, as it only includes effects of freezing on anthropogenic black carbon, while the simulation MIX-nBC also excluded freezing on natural black carbon particles.

### 5.4 Discussion and conclusions

In the global climate model ECHAM5-HAM, we take the dependence of heterogeneous freezing on the composition of aerosol particles available for ice nucleation into account. Two types of mineral dust, montmorillonite and kaolinite, and black carbon are considered as ice nuclei. Uncoated particles can
Figure 5.12: Difference between present-day and preindustrial simulations for the zonal annual means of (a) aerosol optical depth (AOD), (b) cloud cover, (c) liquid water path (LWP) and (d) top-of-the-atmosphere (TOA) short wave (SW) radiation for the experiments MIX and MIX-nBC (without freezing on black carbon).

Table 5.5: Hemispheric annual mean difference in liquid water path (LWP), ice water path (IWP), cloud droplet number burden ($N_B^l$), ice crystal number burden ($N_B^i$), cloud cover (CC) and top-of-the-atmosphere shortwave ($F_{SW}$), longwave ($F_{LW}$) and net ($F_{net}$) radiative fluxes between present-day and preindustrial simulations for the Northern (NH) and Southern Hemisphere (SH) in simulations MIX and MIX-nBC. $F_{SW}$ and $F_{net}$ are upward positive, $F_{LW}$ is downward positive.

<table>
<thead>
<tr>
<th>Simulation</th>
<th>NH</th>
<th>MIX</th>
<th>NH</th>
<th>MIX</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta$LWP [g m$^{-2}$]</td>
<td>11.44</td>
<td>12.01</td>
<td>3.07</td>
<td>2.07</td>
</tr>
<tr>
<td>$\Delta$IWP [g m$^{-2}$]</td>
<td>0.40</td>
<td>0.61</td>
<td>0.01</td>
<td>0.19</td>
</tr>
<tr>
<td>$\Delta N_B^l [10^{10} \text{ m}^{-2}]$</td>
<td>1.74</td>
<td>1.82</td>
<td>0.39</td>
<td>0.32</td>
</tr>
<tr>
<td>$\Delta N_B^i [10^{10} \text{ m}^{-2}]$</td>
<td>0.16</td>
<td>0.12</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>$\Delta$CC [%]</td>
<td>0.87</td>
<td>0.97</td>
<td>0.09</td>
<td>0.01</td>
</tr>
<tr>
<td>$\Delta F_{SW}$ in [W m$^{-2}$]</td>
<td>3.69</td>
<td>3.80</td>
<td>0.81</td>
<td>0.46</td>
</tr>
<tr>
<td>$\Delta F_{LW}$ in [W m$^{-2}$]</td>
<td>0.38</td>
<td>0.60</td>
<td>+0.02</td>
<td>+0.14</td>
</tr>
<tr>
<td>$\Delta F_{net}$ in [W m$^{-2}$]</td>
<td>3.31</td>
<td>3.20</td>
<td>0.78</td>
<td>0.60</td>
</tr>
</tbody>
</table>
initiate contact freezing, while coated particles are potential immersion nuclei, which are active at lower temperatures.

In addition to sensitivity simulations which assume all dust to act as kaolinite/montmorillonite ice nuclei, we present simulations with a three-dimensional atmospheric dust mineralogical composition, which is calculated off-line. The simulated dust composition is more uniform than point measurements of atmospheric dust samples from different continents. In general, the kaolinite content seems to be overestimated.

In our simulations, contact freezing by black carbon is the dominating freezing process, due to high black carbon number concentrations, and due to the small particle size and high Brownian collision efficiency of black carbon. Freezing on dust occurs in both the contact and immersion freezing modes if the dust is composed of montmorillonite (a highly efficient ice nucleus), and almost exclusively in the contact mode if the dust is composed of kaolinite (a less efficient ice nucleus). In present-day climate, anthropogenic sulfate emissions lead to enhanced coating of dust particles, and therefore quasi-deactivate kaolinite ice nuclei. This deactivation effect counteracts the glaciation indirect effect, which suggests an increase of cloud glaciation and precipitation formation resulting from the increase of black carbon ice nuclei resulting from anthropogenic activity (Figure 5.14). The coating and deactivation of natural IN leads to less frequent glaciation of mixed-phase clouds, therefore less precipitation via the ice phase, longer cloud lifetimes, and increased cloud cover and a higher global cloud albedo, resulting in an increase in reflected solar radiation. The magnitude of the deactivation effect depends on the freezing efficiency of dust and thus on the mineralogical composition.

In general, we find a smaller effect of the different dust compositions than a previous study with the ECHAM4 model by Lohmann and Diehl (2006). First of all, the coating and deactivation of dust ice nuclei by anthropogenic sulfate, which counteracts the glaciation indirect effect, was not taken into account in ECHAM4. Furthermore, in ECHAM5-HAM, heterogeneous freezing acts as a trigger to initiate the Bergeron-Findeisen process, which rapidly glaciates clouds. In ECHAM4, the Bergeron-Findeisen process could last several time steps. Although the Bergeron-Findeisen process in ECHAM5-HAM is described as a simple instantaneous threshold process, which might mask the effects of different ice nuclei concentrations, it results in a reasonable partitioning between the liquid and ice phase (Lohmann et al., 2007) and is a considerable improvement over ECHAM4. A more sophisticated parameterization of the Bergeron-Findeisen process, taking into account time-dependent liquid-to-ice conversion as a function of vertical velocities and ice crystal numbers, could enhance the difference between the sensitivity simulations in ECHAM5-HAM.

Finally, we present a sensitivity study with suppressed freezing on black carbon. The glaciation indirect effect is only apparent on the Northern Hemisphere, where heterogeneous freezing is largest. Here it amounts to a reduction in reflected shortwave radiation at top-of-the-atmosphere of \(+0.11\) W m\(^{-2}\) and an increase in longwave radiation emitted to space of \(-0.22\) W m\(^{-2}\). On the Southern Hemisphere, the glaciation indirect effect is masked by changes in sea salt emissions due to variation in surface winds.

We conclude that coating and deactivation of ice nuclei has to be considered next to the emission of additional ice nuclei by anthropogenic activity for estimation of the anthropogenic indirect aerosol effect on
5.4 Discussion and conclusions

Figure 5.14: Schematic of aerosol indirect effects in warm and mixed-phase clouds. The glaciation indirect has
been introduced by Lohmann (2002a). The ‘deactivation indirect effect’ proposed in this study is strongest for
dust composed of a mineral which is inefficient in initiating freezing in the immersion mode (like kaolinite).

mixed-phase clouds. As these two effects counteract each other, the total anthropogenic effect on mixed-
phase clouds is small compared to the aerosol indirect effect on warm clouds. As uncertainties regarding
the parameterization of mixed-phase cloud processes (heterogeneous freezing, Bergeron-Findeisen process)
persist, further field observations of ice nuclei concentrations, composition and related cloud properties
are needed in order to permit model validation.

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program NCCR Climate.
Dust and the glaciation indirect effect
6 Summary and outlook

6.1 Summary

This thesis covers some aspects of the treatment of cloud effects on aerosols, and aerosol effects on clouds, in a global climate model. The focus is on stratiform mixed-phase clouds. Cloud effects on aerosols, the so-called “aerosol processing”, are represented in a newly developed extension of the aerosol module HAM with two new separate modes for in-droplet and in-crystal particles. Aerosol particles are incorporated into droplets through activation and collisions and are released again during droplet evaporation. The re-emission from evaporating droplets is important in mixed-phase clouds, when glaciation via the Wegener-Bergeron-Findeisen-process occurs. Comparison of single-column model simulations to observations of cloud and aerosol parameters at a high-altitude research station in the Swiss Alps shows that the uptake and release processes are generally reasonably simulated, but cloud glaciation is overestimated. Applied on the global scale, the new aerosol processing scheme allows the evaluation of the uptake, release and removal processes of different aerosol species into the liquid and ice phase of clouds. Collision scavenging is important for small particles, while nucleation scavenging and wet removal dominates for example, for sea salt. The global aerosol distribution is substantially modified with the new treatment, and cloud microphysical parameterizations have to be adjusted. Contrary to previous estimates, we find from our simulations that only a relatively small fraction of in-cloud particles is released again to the interstitial phase, and that on average, sampled at a remote location, less than every second particle has already undergone a cloud cycle. The prognostic treatment of the scavenged aerosol fraction is a clear improvement over the use of prescribed scavenging ratios, especially below 0°C, as can be shown in comparison to observations.

In a model intercomparison study for two cases of Arctic stratus clouds, the model’s ability to simulate the development and vertical structure of mixed-phase clouds is demonstrated. The results are very sensitive to the prescribed aerosol parameters. In a set of global simulations, new heterogeneous freezing parameterizations are implemented in the ECHAM5-HAM global climate model. The ability of aerosol particles to act as ice nuclei is dependent on their composition and size. Mineral dust, a natural ice nucleus, and black carbon, mainly anthropogenic, compete for ice formation via contact and immersion freezing. We find that the anthropogenic influence on heterogeneous ice nucleation can either consist of an increase, because of emission of black carbon particles, or of a decrease, because highly efficient dry dust particles can become coated and quasi-deactivated by anthropogenic soluble material. The magnitude of the deactivation depends on the mineralogy of the dust particles.

6.2 Future work

In this section, some ideas and starting points for future work are outlined, which extend the studies presented in this thesis.

6.2.1 Collision scavenging

Collision scavenging of aerosol particles in clouds has been treated in a simplified way in chapters 2 and 3. The collision kernels used in this study are based on theoretical considerations by Young (1974b) and are strictly valid only for droplets and aerosol particles of the specified sizes and at fixed atmospheric conditions (temperature and relative humidity). The collision kernel is strongly dependent on droplet and aerosol particle size. As the cloud droplet radius in ECHAM5-HAM can vary over one order of magnitude, from 5 to 50 µm, and the aerosol particle sizes can also vary within the ranges defined for the seven modes, a more accurate determination of the collision kernels is desirable. Such calculations are provided by Croft et al. (in prep. a) as lookup-tables for in-cloud collision scavenging. Furthermore, Croft et al. (in prep. b) have also introduced size-dependent collision parameters for below-cloud scavenging. For some aerosol species, especially sea salt, the new treatment increased below-cloud scavenging by a factor of five.
6.2.2 Aerosol processing in convective clouds

So far, aerosol processing has been treated in ECHAM5-HAM for stratiform clouds only, because the prognostic cloud droplet and ice crystal numbers refer to long-lived, large-scale clouds only. Convective clouds were treated with a mass-flux scheme based on Tiedtke (1989), without calculation of cloud microphysics.

In a recent study, Lohmann (2007) has introduced a double-moment scheme for cloud water and ice in convective clouds, which permits an extension of the aerosol processing scheme to short-lived clouds. Aerosol processing in convective clouds has been treated in previous studies with cloud-resolving models (e.g., Respondek et al., 1995; Ekman et al., 2006). The global estimates of aerosol turnover in clouds by Pruppacher and Jaenicke (1995) suggest that convective clouds contribute significantly to the modification of aerosol properties, because they frequently evaporate and re-release aerosol particles into the atmosphere. A difficulty for the extension of the aerosol processing scheme arises from the fact that so far the cloud cover and the lifetime of convective clouds are not specified.

6.2.3 Coupling freezing parameterizations to the aerosol processing scheme

The explicit representation of droplet aerosol-collision and in-droplet particles, as introduced in chapter 2, offers the possibility for a more physical parameterization of heterogeneous freezing. In chapter 5, freezing efficiencies of the different possible ice nuclei had to be weighted with their relative contribution to the total soluble and insoluble aerosol, respectively. With the explicit calculation of the collision rates and in-droplet aerosol concentrations, the freezing rates can be scaled in relation to the experimental conditions from which the freezing temperatures have been derived. The theoretical framework for these new parameterizations is developed below.

Aerosol-droplet collisions leading to contact freezing

Contact freezing occurs upon collision of an unfrozen droplet with a particle which can potentially induce ice nucleation at the given temperature. The freezing itself is still a stochastic process. The original formulation by Diehl et al. (2006) for contact freezing by different substances has the following form:

\[ N_f = \text{Max}(1, \text{Min}(0, (-a_x(T - 273.15K) - b_x))) N_0 \]  

(6.1)

for different substances \( x \). Here \( N_f \) is the number of frozen droplets, and \( N_0 \) the “total drop number” of particles “colliding with an insoluble particle”. Therefore the rate of change in \( N_f \) for a given temperature \( T \), equivalent to the contact freezing rate, can be related to the rate of droplets colliding with possibly ice nucleating particles:

\[ \frac{dN_f}{dt} = \sum_x \text{Max}(1, \text{Min}(0, (-a_x(T - 273.15K) - b_x))) \frac{dN_0}{dt} \]

\[ \equiv \sum_x \text{Max}(1, \text{Min}(0, (-a_x(T - 273.15K) - b_x))) Q_{\text{coll},x} \]  

(6.2)

This is a more direct application of Diehl et al.’s (2006) parameterization than equations (5.1) and (5.2), but it requires an assumption for the collision rates \( Q_{\text{coll},x} \) of droplets with ice nuclei of substance \( x \). In equation (5.1) this was provided through inserting the number of ice nuclei into Young’s (1974a) parameterization. The new scheme for aerosol processing in clouds (chapter 2) includes a parameterization of the collision rate (equation (2.11)), which can be used here:

\[ Q_{\text{coll},x} = K_{l,x}N_xN_l \]  

(6.3)

\( x \) refers to the possible ice nuclei, uncoated black carbon and uncoated mineral dust (now accumulation and coarse mode dust have to be distinguished, see Table 5.2). \( K_{l,x} \) can either be taken from Table 2.1 or can be calculated by integrating over the actual aerosol and cloud droplet size distributions as described by Croft et al. (in prep. a).

This approach for the contact freezing parameterization is more physical, as it depends on the absolute concentrations of possible ice nuclei and not only of their fractional contributions to the total insoluble aerosol, as in equation (5.1). It corresponds to the application of the same parameterization in parcel model studies (Diehl et al., 2006, 2007).
In-droplet particles causing immersion freezing

The original formulation of immersion freezing by Diehl and Wurzler (2004) has the following form:

\[
Q_{\text{frz,imm}} \equiv -\frac{dN_i}{dt}_{\text{frz,imm}} = N_i V_d c_x \exp(273.15 - T) \frac{dT}{dt} \quad \text{for} \quad \frac{dT}{dt} < 0 \tag{6.4}
\]

It has been applied by Diehl and Wurzler (2004) for the case of all ice nucleating aerosols consisting of the same material; therefore no weighting of different substances was required. The droplet volume \( V_d \) in the previous expression can be expressed as \( \rho d^3 / 6 \), compare equation (5.6).

As the ice nucleating efficiency in the immersion mode, \( c_x \), has been derived from different laboratory studies with highly concentrated dispersions of ice nucleating particles, it seems desirable to account for this concentration. This can be done in the following way:

\[
Q_{\text{frz,imm}} = \sum_x c_x \frac{N_x \text{in droplet}}{N_x \text{in droplet, exp}} \frac{q_l}{\rho_l} \exp(273.15 - T) \frac{dT}{dt} \quad \text{for} \quad \frac{dT}{dt} < 0 \tag{6.5}
\]

Here \( N_x \text{in droplet} \) is the concentration of particles of substance \( x \) which are suspended in the supercooled droplets for which freezing is to be calculated, and \( N_x \text{in droplet, exp} \) the respective concentrations in the experiments from which the parameters \( c_x \) have been derived. \( N_x \text{in droplet} \) has the dimension [number of particles/volume of liquid]. Unfortunately, \( N_x \text{in droplet, exp} \) is not given by the authors of the experimental studies, but in some cases it can be estimated from the information given in the articles (Table 6.1). The calculated values range between \( 10^8 \) and \( 10^{17} \text{m}^{-3} \). It would be desirable to obtain order-of-magnitude estimates for the studies in which the necessary information was not given (notably for the studies on black carbon ice nuclei (Diehl and Mitra, 1998) and on different clays (Hoffer, 1961)), possibly by repeating the procedure of sample preparation.

With the newly implemented aerosol processing scheme, the average number of possible ice nuclei in cloud droplets \( N_{x, \text{in droplet}} \) can be calculated. The in-droplet mode is considered internally mixed with only one particle per droplet, but by dividing the in-droplet black carbon and dust masses, respectively, by the average mass of the interstitial externally mixed black carbon and dust particles, an approximate value for \( N_{x, \text{in droplet}} \) can be obtained. Compared to equation (5.7), this new formulation has the advantage that the absolute number concentration and not only the fractional composition of the in-cloud aerosol determines immersion freezing. Furthermore, particles from the insoluble modes are counted as possible immersion nuclei, if they have been scavenged by the droplets via collisions, in addition to activated particles from the soluble/mixed modes.

**Expected influence on in-droplet and in-crystal particles**

With a freezing parameterization depending on the concentration of uncoated dust and black carbon particles in the interstitial air and on the concentration of dust and black carbon particles suspended in the droplets, the simulated composition of the in-crystal particles will reflect the contributions of different ice nuclei. It is expected that in-crystal particles will be enriched in black carbon and mineral...
Summary and outlook

dust compared to the interstitial aerosol. Such an enrichment in black carbon has been observed at the Jungfraujoch (Cozic, 2007). Also the probability of cloud glaciation via (contact) freezing at high subzero temperatures can be better evaluated when using a more accurate description of the collision probability. E.g. Sassen et al. (2003) report the glaciation of a mildly supercooled (−5° to −9°C) cloud in the presence of mineral dust aerosol. We will be able to address the question how frequent such events are on the global scale, and whether their occurrence is influenced by anthropogenic aerosols via either the glaciation or the deactivation indirect effect.

Furthermore, depletion of ice nuclei is directly included in the new description, as aerosol particles inside frozen droplets are transferred to the in-crystal mode and do not contribute to the number of potential ice nuclei any longer. Ice nuclei depletion has been shown to be important for persisting mixed-phase clouds (Morrison et al., 2005).

6.2.4 Biogenic ice nuclei

Already 30 years ago, it was recognized in laboratory experiments that certain biogenic materials can nucleate ice at high subzero temperatures (Schnell and Vali, 1976). Pollen (Diehl et al., 2002), fungi, plant debris (Schnell and Vali, 1972; Diehl et al., 2006) and especially bacteria (Vali et al., 1976; Levin and Yankośky, 1983; Morris et al., 2004) can initiate ice formation at temperatures up to −2°C (see the review by Szyrmer and Zawadzki, 1997). Repeatedly the question has been raised how important material derived from living organisms can be for atmospheric ice formation. In-situ detection of organic ice nuclei is difficult, because common methods like aerosol mass spectroscopy do not provide discrimination of different organic materials, which have very similar chemical compositions. GCM simulations with suitable freezing parameterizations could contribute to answering this question, but the concentrations of biogenic aerosols in the atmosphere also need to be simulated adequately. Bioaerosols, of sizes ranging between 10 nm and 100 µm (Ariya and Amyot, 2004), have highly variable emission strengths, atmospheric lifetimes and number concentrations. Helbig et al. (2004) formulated an emission function of pollen as a function of windspeed and different plant parameters for a mesoscale model. Suitable source formulations for other biogenic aerosol particles and for global models are still to be developed. An additional difficulty is the global availability of the required vegetation parameters.

Including biogenic ice nuclei is a desirable future extension of ECHAM5-HAM. Especially bacteria, which are small enough to remain aloft for longer times and can efficiently initiate ice formation, are expected to compete to some extent with mineral dust and black carbon. Preferably the above-described new treatment of contact and immersion freezing should be used, as it directly depends on the ice nuclei number concentrations.
### List of symbols and acronyms

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a_x$</td>
<td>K$^{-1}$</td>
<td>constant for contact freezing (see table 5.1)</td>
</tr>
<tr>
<td>ABS</td>
<td>$-$</td>
<td>absorption aerosol optical depth</td>
</tr>
<tr>
<td>AI</td>
<td>$b$</td>
<td>accumulation insoluble mode</td>
</tr>
<tr>
<td>AOD</td>
<td>$-$</td>
<td>aerosol optical depth</td>
</tr>
<tr>
<td>AP</td>
<td>$-$</td>
<td>simulation with the new aerosol processing scheme</td>
</tr>
<tr>
<td>AS</td>
<td>$-$</td>
<td>accumulation soluble/mixed mode</td>
</tr>
<tr>
<td>$b$</td>
<td>$%$</td>
<td>cloud fraction per gridbox in each vertical level</td>
</tr>
<tr>
<td>$b_{abs}$</td>
<td>m$^{-1}$</td>
<td>aerosol absorption coefficient</td>
</tr>
<tr>
<td>$b_{ext}$</td>
<td>m$^{-1}$</td>
<td>aerosol extinction coefficient</td>
</tr>
<tr>
<td>$b_{scat}$</td>
<td>m$^{-1}$</td>
<td>aerosol scattering coefficient</td>
</tr>
<tr>
<td>$b_x$</td>
<td>$-$</td>
<td>constant for contact freezing (see table 5.1)</td>
</tr>
<tr>
<td>BC</td>
<td>$-$</td>
<td>black carbon</td>
</tr>
<tr>
<td>$C_{AP,c}$</td>
<td>Tg yr$^{-1}$</td>
<td>globally integrated rate of aerosol mass transferred into clouds</td>
</tr>
<tr>
<td>$c_p$</td>
<td>J kg$^{-1}$ K$^{-1}$</td>
<td>specific heat of air at constant pressure</td>
</tr>
<tr>
<td>$C_{v,c}$</td>
<td>g m$^{-2}$ h$^{-1}$</td>
<td>vertically integrated rate of condensation of water vapor mass</td>
</tr>
<tr>
<td>$c_x$</td>
<td>m$^{-3}$</td>
<td>constant for immersion freezing (see table 5.1)</td>
</tr>
<tr>
<td>$CC$</td>
<td>$%$</td>
<td>cloud cover</td>
</tr>
<tr>
<td>CCN</td>
<td>$-$</td>
<td>cloud condensation nuclei</td>
</tr>
<tr>
<td>CD</td>
<td>$-$</td>
<td>in-droplet mode</td>
</tr>
<tr>
<td>CI</td>
<td>$-$</td>
<td>coarse insoluble mode</td>
</tr>
<tr>
<td>cnt</td>
<td>$-$</td>
<td>contact freezing</td>
</tr>
<tr>
<td>CPI</td>
<td>$-$</td>
<td>cloud particle imager</td>
</tr>
<tr>
<td>CS</td>
<td>$-$</td>
<td>coarse soluble/mixed mode</td>
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<tr>
<td>CTL</td>
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<td>control simulation</td>
</tr>
<tr>
<td>CTM</td>
<td>$-$</td>
<td>chemical transport model</td>
</tr>
<tr>
<td>$D_{ap}$</td>
<td>cm$^2$ s$^{-1}$</td>
<td>aerosol diffusivity</td>
</tr>
<tr>
<td>DU</td>
<td>$-$</td>
<td>mineral dust</td>
</tr>
<tr>
<td>DUST</td>
<td>$-$</td>
<td>mineral dust</td>
</tr>
<tr>
<td>$E_{AP,c}$</td>
<td>Tg yr$^{-1}$</td>
<td>globally integrated rate of aerosol mass release from evaporating clouds</td>
</tr>
<tr>
<td>$E_{SS}$</td>
<td>$\mu$g m$^{-2}$ s$^{-1}$</td>
<td>sea salt emission flux</td>
</tr>
<tr>
<td>$E_{v,c}$</td>
<td>g m$^{-2}$ h$^{-1}$</td>
<td>vertically integrated rate of evaporation of water mass</td>
</tr>
<tr>
<td>$E_{v,in-cloud}$</td>
<td>g m$^{-2}$ h$^{-1}$</td>
<td>vertically integrated rate of in-cloud evaporation of water mass</td>
</tr>
<tr>
<td>ECHAM</td>
<td>$-$</td>
<td>GCM derived from the ECMWF model, developed at the Max-Planck Institute in Hamburg, Germany</td>
</tr>
<tr>
<td>ECMWF</td>
<td>$-$</td>
<td>European Centre for Medium-Range Weather Forecasts</td>
</tr>
<tr>
<td>$F_{AP}$</td>
<td>$-$</td>
<td>scavenged fraction for simulation AP</td>
</tr>
<tr>
<td>$F_{CTL,1}$</td>
<td>$-$</td>
<td>scavenged fraction for simulation CTL, definition 1</td>
</tr>
<tr>
<td>$F_{CTL,2}$</td>
<td>$-$</td>
<td>scavenged fraction for simulation CTL, definition 2</td>
</tr>
<tr>
<td>$F_{lw}$</td>
<td>W m$^{-2}$</td>
<td>top-of-the-atmosphere longwave radiative flux</td>
</tr>
<tr>
<td>$F_{n}$</td>
<td>$-$</td>
<td>scavenged particle number fraction</td>
</tr>
<tr>
<td>$F_{net}$</td>
<td>W m$^{-2}$</td>
<td>top-of-the-atmosphere net (shortwave+longwave) radiative flux</td>
</tr>
<tr>
<td>$F_{sw}$</td>
<td>W m$^{-2}$</td>
<td>top-of-the-atmosphere shortwave radiative flux</td>
</tr>
<tr>
<td>$g$</td>
<td>m s$^{-2}$</td>
<td>acceleration of gravity</td>
</tr>
</tbody>
</table>

*a $-^a$ indicates a dimensionless variable.

*b No units are given for abbreviations.
List of symbols and acronyms

GAW  Global Atmosphere Watch
GCM  global climate model, general circulation model
HAM  Hamburg Aerosol Module
IC   in-crystal mode
imm  immersion freezing
IN cm$^{-3}$ ice nuclei
IPCC Intergovernmental Panel on Climate Change
IWC g m$^{-3}$ ice water content
IWP g m$^{-2}$ ice water path
JFJ  Jungfraujoch
$K_{i,j}$ m$^3$s$^{-1}$ collision kernel for ice crystals with aerosols of mode $j$
$K_{l,j}$ m$^3$s$^{-1}$ collision kernel for cloud droplet with aerosols of mode $j$
KAO kaolinite
KI  Aitken insoluble mode
KS  Aitken soluble/mixed mode
LCF W m$^{-2}$ longwave cloud forcing
LWC g m$^{-3}$ liquid water content
LWP g m$^{-2}$ liquid water path
$m_{x,j}$ g m$^{-3}$ aerosol mass concentration of component $x$ in mode $j$
MBL marine boundary layer
MON montmorillonite
$N_0$ cm$^{-3}$ initial cloud droplet number concentration
$N_{act,j}$ cm$^{-3}$ number concentration of aerosols activated to cloud droplets
$N_{acti,j}$ cm$^{-3}$ number concentration of aerosols activated to ice crystals
$N_{aer>35nm}$ cm$^{-3}$ number concentration of aerosols in the soluble modes with radius larger than 35 nm
$N_{cnt,x}$ cm$^{-3}$ possible contact freezing nuclei of material $x$
$N_f$ cm$^{-3}$ frozen droplet number concentration
$N_i$ cm$^{-3}$ ice crystal number concentration
$N_B$ m$^{-2}$ ice crystal number burden
$N_{imm,x}$ cm$^{-3}$ possible immersion freezing nuclei of material $x$
$N_{acti}$ cm$^{-3}$ aerosol number concentration, mode $j$
$N_{int}$ cm$^{-3}$ interstitial aerosol number concentration
$N_l$ cm$^{-3}$ cloud droplet number concentration
$N_B$ m$^{-2}$ cloud droplet number burden
$N_{cloudy}$ m$^{-2}$ cloud droplet number burden, averaged over cloudy areas only
$N_{tot}$ cm$^{-3}$ total aerosol number concentration
NCCR National Centre of Competence in Research
NS nucleation soluble mode
OC organic carbon
$P$ mm yr$^{-1}$ precipitation
$p$ hPa pressure
PD present-day
PI preindustrial
POM particulate organic matter
$Q$ cm$^{-3}$ s$^{-1}$ grid-mean rate
$Q_{acc}$ cm$^{-3}$ s$^{-1}$ accretion rate of cloud droplets by falling snow
$Q_{accl}$ cm$^{-3}$ s$^{-1}$ accretion rate of cloud ice crystals by falling snow
$Q_{agg}$ cm$^{-3}$ s$^{-1}$ aggregation rate of ice crystals to snow
$Q_{aut}$ cm$^{-3}$ s$^{-1}$ autoconversion rate of cloud droplets to rain
$Q_{coll}$ cm$^{-3}$ s$^{-1}$ collision rate of aerosols with cloud droplets
$Q_{colli}$ cm$^{-3}$ s$^{-1}$ collision rate of aerosols with ice crystals
$Q_{evap}$ cm$^{-3}$ s$^{-1}$ cloud droplet evaporation rate
$Q_{frz}$ cm$^{-3}$ s$^{-1}$ freezing rate
$Q_{frz,act}$ cm$^{-3}$ s$^{-1}$ contact freezing rate
$Q_{frz,imm}$ cm$^{-3}$ s$^{-1}$ immersion freezing rate
$q_i$ g m$^{-3}$ ice mass concentration
$q_l$ g m$^{-3}$ liquid water mass concentration
\(Q_{\text{melt}}\) cm\(^{-3}\) s\(^{-1}\) melting rate
\(Q_{\text{nuc}}\) cm\(^{-3}\) s\(^{-1}\) cloud droplet nucleation rate
\(Q_{\text{nuci}}\) cm\(^{-3}\) s\(^{-1}\) ice crystal nucleation rate
\(q_{\text{rain}}\) g m\(^{-3}\) rain water mass concentration
\(q_s\) g kg\(^{-1}\) saturation mixing ratio
\(Q_{\text{secp}}\) cm\(^{-3}\) s\(^{-1}\) ice crystal secondary production rate
\(Q_{\text{self}}\) cm\(^{-3}\) s\(^{-1}\) cloud droplet selfcollection rate
\(Q_{\text{selfi}}\) cm\(^{-3}\) s\(^{-1}\) ice crystal selfcollection rate
\(q_{\text{snow}}\) g m\(^{-3}\) snow water mass concentration
\(Q_{\text{sub}}\) cm\(^{-3}\) s\(^{-1}\) ice crystal sublimation rate
\(q_t\) g kg\(^{-1}\) total (vapor + condensed) water mixing ratio
\(R\) – average scavenging coefficient
\(R_j\) – scavenging coefficient for mass and number of mode \(j\)
\(r\) mm, \(\mu\)m number median aerosol radius
\(\bar{r}\) nm, \(\mu\)m number median aerosol radius
\(r_{\text{eff,ice}}\) \(\mu\)m effective ice crystal radius
\(r_{\text{eff,l}}\) \(\mu\)m effective droplet radius
\(r_l\) \(\mu\)m droplet radius
\(S\) sulfur
\(S_{\text{AP}}\) Tg yr\(^{-1}\) global surface source of aerosol particles
SCF W m\(^{-2}\) shortwave cloud forcing
SCM single column model
SMPS scanning mobility particle sizer
SO2 sulfur dioxide
SO4 sulfate
SO4(g) gaseous sulfuric acid
SS sea salt
\(T\) K, °C temperature
\(t\) s time
\(T_d\) K dewpoint temperature
TKE \(m^2\) s\(^{-2}\) turbulent kinetic energy
TOA top of the atmosphere
TWC g m\(^{-3}\) total (liquid+ice) water content
TWP g m\(^{-2}\) total (liquid+ice) water path
UTC coordinated universal time
\(V\) \(\mu\)m\(^3\) \(\mu\)m\(^{-2}\) column integrated volume volume
\(V_d\) \(\mu\)m\(^3\) droplet volume
\(V_j\) \(\mu\)m\(^3\) kg\(^{-1}\) total dry volume of aerosol from mode \(j\) per kg of air
\(w\) m s\(^{-1}\) vertical velocity
\(\bar{w}\) m s\(^{-1}\) large-scale vertical velocity
\(W_{\text{dep}}\) Tg yr\(^{-1}\) global wet deposition rate of aerosol mass
\(W_{\text{dep,bc}}\) Tg yr\(^{-1}\) global below-cloud wet deposition rate of aerosol mass
\(W_{\text{dep,ic}}\) Tg yr\(^{-1}\) global in-cloud wet deposition rate of aerosol mass
\(x_{\text{KAO, MON}}\) mass fractions of kaolinite and montmorillonite
\(z\) m vertical coordinate

**Greek letters**
\(\alpha\) cm\(^{-4}\) s\(^{-1}\) constant in Lin and Leaitch’s (1997) activation parameterization
\(\gamma\) – autoconversion tuning parameter
\(\Delta\) difference
\(\theta\) K potential temperature
\(\lambda\) nm wavelength
\(\rho_{\text{air}}\) kg m\(^{-3}\) air density
\(\rho_j\) kg m\(^{-3}\) density of mode/component \(j\)
\(\rho_l\) kg m\(^{-3}\) liquid water density
\(\sigma_j\) – standard deviation of mode \(j\)
\(\tau_{c,\text{evap}}\) min cloud lifetime with respect to evaporation
\(\tau_{c,\text{precip}}\) h cloud lifetime with respect to precipitation
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Curriculum vitae

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Conferences, workshops and summer schools

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HAM User Workshop, Hamburg, Germany (2006)


EGU General Assembly, Vienna, Austria (2006)


IAMAS, Beijing, China (2005)

4th International NCCR Climate Summer School, “From the Holocene to the Anthropocene: Climate of the last 1000 Years”, Grindelwald, Switzerland (2005)

1st French-German Summer School on “Aerosols and Heterogeneous Chemistry”, Ile d’Oléron, France (2004)

DACH Meteorologen-Tagung, Karlsruhe, Germany (2004)
Curriculum vitae

Publications


Hoose, C. (2003), ‘Modélisation numérique du cycle du soufre atmosphérique en climat glaciaire (Numerical modeling of the atmospheric sulfur cycle in glacial climate)’, Rapport de Stage Maîtrise, Université Joseph Fourier, Grenoble (report on a 3-months research project).
Danke!