Sensitivity Studies of Aerosol–Cloud Interactions in Mixed-Phase Orographic Precipitation

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ABSTRACT

Anthropogenic aerosols serve as a source of both cloud condensation nuclei (CCN) and ice nuclei (IN) and affect microphysical properties of clouds. Increasing aerosol number concentration is assumed to retard the cloud droplet coalescence and the riming process in mixed-phase orographic clouds, thereby decreasing orographic precipitation.

In this study, idealized 3D simulations are conducted to investigate aerosol–cloud interactions in mixed-phase orographic clouds and the possible impact of anthropogenic and natural aerosols on orographic precipitation. Two different types of aerosol anomalies are considered: naturally occurring mineral dust and anthropogenic black carbon.

In the simulations with a dust aerosol anomaly, the dust aerosols serve as efficient ice nuclei in the contact mode, leading to an early initiation of the ice phase in the orographic cloud. As a consequence, the riming rates in the cloud are increased, leading to increased precipitation efficiency and enhancement of orographic precipitation.

The simulations with an anthropogenic aerosol anomaly suggest that the mixing state of the aerosols plays a crucial role because coating and mixing may cause the aerosols to initiate freezing in the less efficient immersion mode rather than by contact nucleation. It is found that externally mixed black carbon aerosols increase riming in orographic clouds and enhance orographic precipitation. In contrast, internally mixed black carbon aerosols decrease the riming rates, leading in turn to a decrease in orographic precipitation.

1. Introduction

Understanding orographic precipitation is an important aspect for the hydrology, ecology, and the local climate in mountainous regions (Smith et al. 2005). The controlling factors of orographic precipitation found during the Mesoscale Alpine Programme (MAP; Bougeault et al. 2001; Volkert and Gutermann 2007) are attributable to the orography (e.g., geometry of the terrain), the dynamics of the flow (e.g., flow blocking, turbulence), the thermodynamic properties of the air mass (e.g., thermal stratification, convection) and the cloud microphysics (Rotunno and Houze 2007).

Because parts of the atmospheric aerosol spectrum serve as cloud condensation nuclei (CCN) and ice nuclei (IN), these particles play a crucial role in controlling the microphysical properties of orographic clouds and potentially affect the orographic precipitation development. An increase in the CCN shifts the cloud droplet size spectrum toward smaller radii and reduces the efficiency of the collision/coalescence process in warm-phase clouds (e.g., Twomey et al. 1984; Peng et al. 2002; Lowenthal et al. 2004).

For mixed-phase orographic clouds, observations by Borys et al. (2000, 2003) suggest a tendency of decreasing snowfall rate with increasing anthropogenic aerosol load due to a suppression of the collection of cloud droplets by snowflakes, which is subsequently referred to as riming. Because collision and riming are microphysical processes for producing precipitation efficiently, an inhibition of these cloud microphysical processes is assumed to yield a prolongation of the precipitation
development. This delay in the production of precipitable hydrometeors may affect the orographic precipitation distribution and the total orographic precipitation budgets (e.g., Hobbs et al. 1973).

Another aspect of aerosol–cloud interactions in mixed-phase orographic clouds arises from the ice-nucleating abilities of different aerosol types. Aerosols may serve as IN and hence are responsible for initiating the ice phase in cold clouds via heterogeneous ice nucleation. The efficiency of aerosols to act as heterogeneous IN depends on their chemical composition as recently reviewed by Baker and Peter (2008) but also on their surface properties (e.g., crystallographic structure of the lattice, surface defects, active sites; Pruppacher and Klett 1997; Marcolli et al. 2007; Zobrist et al. 2008). Heterogeneous ice nucleation is the dominant nucleation process in most tropospheric clouds (Cantrell and Heymsfield 2005) and is therefore also important for low-level orographic clouds. Here, it is important to note that different modes of ice nucleation exist that are dependent on the ambient temperature and supersaturation with respect to ice (Pruppacher and Klett 1997; Vali 1985).

The overall implications of the indirect aerosol effect on orographic clouds and precipitation at different spatial and temporal scales are inconclusive and uncertain for both observational and modeling studies (Denman et al. 2007; Levin and Cotton 2008). For example, some statistical analyses of rain gauge data in the United States, China, and Israel suggest a suppression of orographic precipitation with increasing aerosol load (Givati and Rosenfeld 2004; Jirak and Cotton 2006; Rosenfeld et al. 2007; Givati and Rosenfeld 2007), whereas findings by Alpert et al. (2008) contradict these results. Lohmann (2002) conducted global model simulations and concluded that the availability of contact nuclei such as dust or soot may lead to enhanced glaciation of clouds (the so-called glaciation indirect effect) and to an increase in precipitation via the ice phase. In contrast, Lohmann and Zhang (2003) simulated Arctic clouds and found that an increase in the aerosol load leads to a decrease in the overall precipitation. However, Lohmann and Zhang (2003) also found that the amount of snowfall critically depends on the snow crystal geometry. If planar crystals were assumed, the increase in the aerosol led to a decrease in snowfall whereas for aggregates an increase in snowfall was found. Furthermore, Lohmann (2004) observed a reduction in the riming rate if a size-dependent collection efficiency for snowflakes was introduced in the model, which corroborates findings by Borys et al. (2000, 2003). Recently, 2D simulations by Lynn et al. (2007) on mixed-phase orographic clouds suggested a decrease of orographic precipitation if the background aerosol conditions changed from maritime to continental. However, confident estimates for the possible implications of the aerosol–cloud interactions on the mixed-phase precipitation formation are lacking because of the uncertainties in representing microphysical processes in numerical models (e.g., heterogeneous ice nucleation) and the scarceness of observational data (e.g., Levin and Cotton 2008).

The main goal of this paper is to investigate how natural as well as anthropogenic aerosol perturbations may affect microphysical processes in mixed-phase (stratiform) orographic clouds and to what extent aerosol–cloud interactions may change orographic precipitation (i.e., amount and distribution). Therefore, we address the following questions:

- Is the proposed mechanism of inhibited riming reproducible with a state-of-the-art mesoscale weather prediction model?
- How strongly may the orographic precipitation pattern and the orographic precipitation budget be affected by natural and anthropogenic aerosol perturbations?
- How sensitive is orographic precipitation to the freezing efficiencies of different aerosols types at different temperatures?

The paper is structured as follows: In section 2 we introduce the modeling approach focusing on the numerical model and the parameterizations that are employed. In section 3 we discuss the model setup and the experimental design. In sections 4 and 5 we conduct different sensitivity experiments to investigate the influence of different aerosol configurations on orographic precipitation at different temperatures. Finally, we end with a discussion of our results and the conclusions in section 6.

2. Numerical model

The model simulations are performed with the non-hydrostatic, fully compressible, limited-area mesoscale weather prediction Consortium for Small-Scale Modeling (COSMO) model (formerly known as the LM; Doms and Schätttler 2002; Steppeler et al. 2003; http://www.cosmo-model.org). The elastic equations are solved in a split-explicit time-splitting approach (Wicker and Skamarock 2002) with a two time-level total variation diminishing (TVD) third-order Runge–Kutta scheme in combination with a fifth-order horizontal advection scheme. All moisture variables and aerosols are advected by a fourth-order positive-definite advection scheme after Bott (1989).
The boundary conditions are open at the lateral boundaries (Davies 1976) and free-slip at the lower boundary. A Rayleigh damping sponge layer is introduced at the upper rigid boundary to damp reflections of vertically propagating gravity waves. The damping layer starts at 10 km height and covers approximately half of the vertical model domain.

Because the main focus of this study is on aerosol–cloud–precipitation interactions via the aerosol indirect effect, all radiative effects such as the change in cloud albedo are neglected. Thus, no parameterization of radiative processes is considered here. Furthermore, the convection parameterization is switched off.

For the vertically turbulent diffusive processes, a 2.5-level Mellor–Yamada scheme with a prognostic turbulent kinetic energy (TKE) equation is used (Herzog et al. 2002).

The coupled cloud-microphysical and aerosol-microphysical processes are treated in a two-moment approach and are discussed next.

a. Aerosol microphysics

The aerosol-microphysics scheme is based on the aerosol module M7 (Vignati et al. 2004; Stier et al. 2005) and treats the aerosol size distribution \( f(\ln r) \) as a superposition of seven lognormal aerosol modes such that

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

Here, \( N_k \) is the aerosol number density \((\text{m}^{-3})\) and \( \bar{r}_k \) (m) is the count median radius of the \( k \)th aerosol mode. The geometric standard deviation \( \sigma_k \) is a free parameter and is kept fixed for each aerosol mode. Furthermore, the aerosol scheme distinguishes between different aerosol species, namely sulfates (SU), carbonaceous aerosols [black carbon (BC) and organic carbon (OC)], sea salt (SS), and mineral dust (DU). The seven aerosol modes are comprised of either soluble/mixed modes (i.e., sulfate, sea salt, mixed black/organic carbon) or insoluble modes (i.e., freshly formed black/organic carbon or mineral dust) and distinguished by size as nucleation (NUC), Aitken (AIT), accumulation (ACC) and coarse (COA) modes. The prognostic variables are the number densities \( N_k \) and the mass densities \( m_{k,j} \) for each mode \( k \) and aerosol species \( j \) (e.g., \( m_{\text{AIT insol.},BC} \) denotes the mass density of insoluble black carbon aerosols in the Aitken mode). The count median radius \( \bar{r}_k \) can then be diagnosed from the prognostic number density \( N_k \) and the prognostic mass densities \( m_{k,j} \) such that

\[
\bar{r}_k = \left( \frac{3}{4\pi N_k} \sum_j m_{k,j} \right)^{1/3} \exp\left(\frac{3}{2} \ln^2 \sigma_k\right),
\]

for all aerosol modes \((k \in \{\text{NUC, AIT, ACC, COA}\})\), aerosol species \((j \in \{\text{SU, BC, OC, SS, DU}\})\) and aerosol densities \( \rho_j \). The aerosol-microphysical processes considered in the model are the nucleation of gas-phase sulfuric acid (Vehkamäki et al. 2002), the condensation of sulfuric acid on pre-existing aerosol particles, the coating of insoluble aerosols by sulfuric acid, inter- and intramodal coagulation, and the uptake of water vapor.

b. Cloud microphysics

The cloud-microphysics parameterization is based on Seifert and Beheng (2006) and considers five hydrometeor categories: cloud droplets (c), rain (r), ice crystals (i), snow (s), and graupel (g). The warm-phase processes of the scheme include the nucleation of cloud droplets, condensation/evaporation of cloud droplets, autoconversion of cloud droplets to rain (i.e., the collision of cloud droplets leading to rain drops), accretion of cloud droplets by rain (i.e., the collection of cloud droplets by raindrops), self-collection of cloud droplets and rain, evaporation of rain, and the breakup of large raindrops as discussed in Seifert and Beheng (2006) and Muhlbauer and Lohmann (2008).

The ice-phase processes include heterogeneous freezing, melting, vapor deposition, sublimation, riming, aggregation, collection, and conversion to graupel. Furthermore, secondary ice nucleation processes such as the Hallett–Mossop ice multiplication are parameterized. We account for heterogeneous freezing nucleation in the immersion and in the contact mode but neglect deposition nucleation and homogeneous ice nucleation. In the present study, condensation freezing is implicitly included in the immersion freezing parameterization and immersion freezing is used as a surrogate for both processes as done previously by Diehl et al. (2006) and Lohmann and Diehl (2006). Laboratory studies by Schaller and Fukuta (1979) suggest that deposition nucleation requires temperatures of approximately \(-20^\circ\text{C}\) and supersaturations with respect to ice of about 20%. At lower supersaturations the onset temperature for deposition nucleation is even lower. Based on studies by Korolev and Mazin (2003) and Korolev and Isaac (2006), the necessary supersaturations for deposition nucleation to occur are unlikely to be maintained in low-level orographic clouds. Hence, we consider deposition nucleation as negligible. Homogeneous ice nucleation requires temperatures below \(-38^\circ\text{C}\) and, thus, is only
Table 1. The coefficients for the generalized gamma distributions and for the power-law relations of the maximum diameter $D(m)$ and the terminal fall velocity $v(m)$ of hydrometeors with mass $m$ used in the cloud-microphysics scheme.

<table>
<thead>
<tr>
<th>Hydrometeor class</th>
<th>$\nu$</th>
<th>$\mu$</th>
<th>$\alpha$ (m kg$^{-b}$)</th>
<th>$b$</th>
<th>$\alpha$ (m s$^{-1}$ kg$^{-b}$)</th>
<th>$\beta$</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cloud droplets (c)</td>
<td>1</td>
<td>1</td>
<td>0.124</td>
<td>1/3</td>
<td>$3.75 \times 10^3$</td>
<td>2/3</td>
<td>1</td>
</tr>
<tr>
<td>Rain (r)</td>
<td>0</td>
<td>1/3</td>
<td>0.124</td>
<td>1/3</td>
<td>159.0</td>
<td>0.266</td>
<td>1/2</td>
</tr>
<tr>
<td>Ice crystals (i)</td>
<td>1/3</td>
<td>-1/3</td>
<td>0.217</td>
<td>0.302</td>
<td>41.9</td>
<td>0.260</td>
<td>1/2</td>
</tr>
<tr>
<td>Snowflakes (s)</td>
<td>1/2</td>
<td>1/2</td>
<td>8.156</td>
<td>0.526</td>
<td>27.7</td>
<td>0.216</td>
<td>1/2</td>
</tr>
<tr>
<td>Graupel (g)</td>
<td>1</td>
<td>1/3</td>
<td>0.190</td>
<td>0.323</td>
<td>40.0</td>
<td>0.230</td>
<td>1/2</td>
</tr>
</tbody>
</table>

Important for cloud formation in the upper troposphere such as in cirrus clouds (Pruppacher and Klett 1997; Szyrmer and Zawadzki 1997; Cantrell and Heymsfield 2005). For the sake of brevity we only discuss the microphysical equations that are most relevant for this study (i.e., the parameterizations of heterogeneous freezing and mixed-phase collections such as riming and aggregation). For a detailed discussion on the complete two-moment microphysics scheme and the cloud droplet activation, see Seifert and Beheng (2006) and Muhlbauer and Lohmann (2008).

All hydrometeor categories treated in the two-moment scheme follow a generalized gamma distribution with respect to the mass such that

$$f_k(m) = A m^{\nu_k} \exp(-\lambda m^\mu_k), \quad k \in \{c, r, i, s, g\}. \quad (3)$$

The free parameters $\mu_k$ and $\nu_k$ of the generalized gamma distribution vary for the different hydrometeor classes and are given in Table 1. The coefficients $A$ and $\lambda$ are related to the zeroth moment $M^0$ and first moment $M^1$ of the underlying hydrometeor size distribution with respect to mass as discussed in Seifert and Beheng (2006). The hydrometeor number densities $N_k$ and the mass densities $L_k$ are defined such that

$$N_k = M^0_k = \int_0^\infty f_k(m) \, dm, \quad (4)$$

$$L_k = M^1_k = \int_0^\infty m f_k(m) \, dm. \quad (5)$$

The ice-phase scheme is in some parts similar to the twomoment scheme described by Reisner et al. (1998). The diameter–mass $D(m)$ and velocity–mass relations $v(m)$ of the different hydrometeors are parameterized depending on the mass $m$ by power laws of the form

$$D(m) = a m^b, \quad (6)$$

$$v(m) = a m^\beta \left( \frac{\rho_0}{\rho} \right)^\gamma, \quad (7)$$

where $\rho$ is the air density and $\rho_0 = 1.225$ kg m$^{-3}$; the coefficients $a$, $b$ and $\alpha$, $\beta$, $\gamma$ are summarized in Table 1. For nonspherical particles $D(m)$ denotes the maximum diameter as discussed in Seifert and Beheng (2006). Note that in the terminal fall velocity Eq. (7) a height correction is applied.

For the geometry and the terminal fall velocities of the different ice-phase hydrometeors, we assume hexagonal plates for the ice crystals (Heymsfield and Kajikawa 1987), mixed aggregates for the snowflakes (Locatelli and Hobbs 1974) and lump graupel with a modified terminal fall velocity (Heymsfield and Kajikawa 1987; Seifert and Beheng 2006).

1) IMMERSION FREEZING

Heterogeneous freezing of cloud droplets in the immersion mode is treated here as a combination of the singular and the stochastic freezing approach. In analogy to homogeneous freezing, heterogeneous freezing is considered to occur in supercooled cloud droplets with the probability of freezing being a function of temperature and droplet volume (Pruppacher and Klett 1997). Aerosols contained in the cloud droplets serve as freezing catalysts and enhance the probability of freezing for a fixed temperature depending on the efficiency of the immersed aerosol to serve as an ice nucleus. Parameterizations of immersion freezing following purely the stochastic hypothesis (Bigg 1953) have been applied in cloud models before (e.g., Reisin et al. 1996; Reisner et al. 1998; Cui et al. 2006). However, the evidence for heterogeneous immersion freezing being solely a stochastic process has been weakening over time as a result of new laboratory measurements (e.g., Vali 1994; Marcolli et al. 2007; Vali 2008). Following Khain et al. (2000) and Seifert and Beheng (2006), the rate equation for heterogeneous immersion freezing for the $k$th moment of a size distribution is

$$\frac{\partial M^k}{\partial t} = -M^{k+1} J(T), \quad (8)$$

where $J(T)$ is the heterogeneous freezing nucleation rate (kg s$^{-1}$ s$^{-1}$). Thus, the rate equations for the number and mass densities can be directly deduced from Eq. (8) for any cloud droplet size distribution. Assuming a gamma distribution for cloud droplets ($\mu = 1$) yields the
following rate equations for the ice-crystal number density \( N_i \) (m\(^{-3}\)) and the mass density \( L_i \) (kg m\(^{-3}\)), respectively:

\[
\frac{\partial N_i}{\partial t}_{\text{IFR}} = -\frac{L_i}{\rho_w} J_{\text{IFR}}(T),
\]

(9)

\[
\frac{\partial L_i}{\partial t}_{\text{IFR}} = \nu_c + 2 \frac{L_i}{\nu_c + 1} \frac{\partial N_i}{\partial t}_{\text{IFR}}.
\]

(10)

Here, \( J_{\text{IFR}}(T) \) (m\(^{-3}\) s\(^{-1}\)) is the heterogeneous nucleation rate for immersion freezing and \( \rho_w \) is the density of water. Because of the conservation of mass and number, we apply \( \frac{\partial N_i}{\partial t}_{\text{IFR}} = -(\frac{\partial L_i}{\partial t})_{\text{IFR}} \) and \( \frac{\partial N_i}{\partial t}_{\text{IFR}} = -(\frac{\partial L_i}{\partial t})_{\text{IFR}} \). The nucleation rate in the immersion mode \( J_{\text{IFR}}(T) \) is parameterized after Diehl and Wurzler (2004) and Lohmann and Diehl (2006)

\[
J_{\text{IFR}}(T) = ab \{ \exp[a(T_0 - T)] - 1 \} \frac{dT}{dt},
\]

(11)

and depends on the temperature \( T \) (K), the Lagrangian cooling rate \( dT/dt \) (K s\(^{-1}\)), the constant \( T_0 = 273.20 \) K, and the parameters \( a \) (K\(^{-1}\)) and \( b \) (m\(^{-3}\)). Equation (11) is only evaluated if the cooling rate \( dT/dt \) is negative, which excludes overnucleation as discussed by Khain et al. (2000). In previous studies such as Reisner et al. (1998), Cotton and Field (2002), or Seifert and Beheng (2006), the parameters \( a \) and \( b \) in Eq. (11) were based on measurements that did not distinguish between different aerosol compositions. However, a series of laboratory experiments and field studies suggest that the chemical composition of aerosols affects their ice-nucleating efficiency and thus is an important aspect that needs to be considered (e.g., Roberts and Hallett 1968; Sassen et al. 2003; DeMott et al. 2004; Cziczo et al. 2004; Cozi et al. 2007a,b). In this work, we explicitly account for the different ice-nucleation efficiencies of aerosols by diagnosing the mean ice-nucleation efficiency \( b \) in Eq. (11) from the size distribution and the chemical composition of aerosols prognosticed by the aerosol microphysics scheme. Thus, we calculate the surface-weighted averaged ice-nucleation efficiency \( b \) such that

\[
b = \frac{\sum B_j S_j}{S}, \quad j \in \{ \text{BC}, \text{DU} \},
\]

(12)

with \( B_j \) being the material-specific but size-independent freezing efficiency of the aerosol component \( j \) and \( S \) the surface area of the immersed aerosol component \( j \) that acts as an ice nucleus. Here, we consider only black carbon and dust as efficient ice nuclei in the immersion mode. The freezing efficiencies \( B_j \) are taken from a compilation of different laboratory studies by Diehl and Wurzler (2004) and are given in Table 2.

### Table 2. Material-specific coefficients for the heterogeneous freezing parameterization. The coefficients are taken from Table 2 of Diehl and Wurzler (2004) for immersion freezing and from Table 3 of Diehl et al. (2006) for contact freezing. Note that for mineral dust the values for montmorillonite are taken.

<table>
<thead>
<tr>
<th>Aerosol species</th>
<th>( a ) (K(^{-1}))</th>
<th>( B ) (m(^{-3}))</th>
<th>( a ) (K(^{-1}))</th>
<th>( b )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Black carbon (BC)</td>
<td>1.0</td>
<td>2.91 \times 10^{-3}</td>
<td>6.14 \times 10^{-2}</td>
<td>57.30 \times 10^{-2}</td>
</tr>
<tr>
<td>Mineral dust (DU)</td>
<td>1.0</td>
<td>32.3</td>
<td>10.14 \times 10^{-2}</td>
<td>32.77 \times 10^{-2}</td>
</tr>
</tbody>
</table>

The parameterization of contact freezing is based on Young (1974), Cotton et al. (1986), and Lohmann and Diehl (2006). We consider contact freezing that is induced by collision of supercooled liquid water droplets and aerosols by Brownian motion. The rate equations for the number density \( N_i \) (m\(^{-3}\) s\(^{-1}\)) and mass density \( L_i \) (kg m\(^{-3}\) s\(^{-1}\)) of the newly forming ice crystals are given by

\[
\frac{\partial N_i}{\partial t}_{\text{CFR}} = 4\pi \bar{r}_c N_c \sum_k D_k N_{k,\text{con}},
\]

(15)

\[
\frac{\partial L_i}{\partial t}_{\text{CFR}} = \frac{L_i}{N_c} \frac{\partial N_i}{\partial t}_{\text{CFR}}.
\]

(16)

Here, \( N_{k,\text{con}} \) is the number density of available contact nuclei in the aerosol mode \( k \) and \( N_c \) is the number density of cloud droplets. The volume mean radius of
cloud droplets, \( \tilde{r}_c \), can be diagnosed from the cloud droplet number concentration \( N_c \) and the cloud droplet mass density \( L_c \) by assuming spherical cloud droplets with density \( \rho_w \):

\[
\tilde{r}_c = \left( \frac{3L_c}{4\pi\rho_w N_c} \right)^{1/3}.
\]

The rate equation [\((15)\)] for contact freezing is limited by the collision between cloud droplets and contact nuclei induced by the Brownian diffusion. Therefore, we consider the aerosol Brownian diffusivity in air \( \text{m}^2 \text{s}^{-1} \),

\[
D_k = \frac{k_B T C}{6\pi \eta f_k},
\]

which depends on the Boltzmann constant \( k_B = 1.38 \times 10^{-23} \text{ m}^2 \text{kg} \text{s}^{-2} \text{K}^{-1} \), the temperature \( T \), the Cunningham slip correction factor \( C \), the viscosity of air \( \eta \) \( \text{kg m}^{-1} \text{s}^{-1} \), and the dry count median radius \( \tilde{r}_k \) of the aerosol mode \( k \). The Cunningham slip correction factor is given by

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

with the molecular mean free path length of air \( \lambda = 0.066 \mu \text{m} \) at the standard conditions \( p_0 = 1013.25 \text{ hPa} \) and \( T_0 = 273.15 \text{ K} \). The viscosity of air is calculated according to

\[
\eta = 10^{-5} \left[ 1.718 + 4.9 \times 10^{-3} (T - T_0) 
- 1.2 \times 10^{-5} (T - T_0)^2 \right].
\]

The aerosol Brownian diffusivity increases with decreasing aerosol radius such that smaller contact nuclei have a higher collision efficiency than larger contact nuclei. At standard conditions the Brownian aerosol diffusivity varies over several orders of magnitude; thus, small contact nuclei such as black carbon can be very efficient solely because of the high collision efficiencies. In the studies by Young (1974) and Cotton et al. (1986) the number of contact nuclei depended explicitly on temperature but the constraint of the limited number of contact nuclei in nature was neglected. To account for this drawback, we limit the number of contact nuclei to the number of insoluble aerosols with the ability to act as contact ice nuclei. Hence, the considered aerosols need to be hydrophobic, nonactivated, and not already be mixed or coated with any water-soluble material. In this study, we follow experimental results by Gorbunov et al. (2001) and Diehl and Wurzler (2004) and consider black carbon and mineral dust aerosols as potential ice nuclei in the contact freezing mode. The number of contact ice nuclei \( N_{k,\text{con}} \) in the aerosol mode \( k \) is calculated according to

\[
N_{k,\text{con}} = \frac{N_k \sum_j \epsilon_j S_j}{S}, \quad j \in \{\text{BC, DU}\}, \tag{21}
\]

with the freezing efficiency \( \epsilon \) calculated as a linear function of temperature

\[
\epsilon_j = a_j T + b_j, \quad 0 \leq \epsilon \leq 1 \tag{22}
\]

with the slope \( a_j \) \( (\text{K}^{-1}) \) and intercept \( b_j \) depending on the aerosol type. The coefficients \( a_j \) and \( b_j \) are based on Diehl et al. (2006) and are given in Table 2.

3) ICE-PHASE COLLECTION/SELF-COLLECTION

For mixed-phase clouds the collection/self-collection of two hydrometeors \( k \) and \( j \) \((k, j \in \{l, s, g\})\) is parameterized in the following way:

\[
\left( \frac{\partial L_k}{\partial t} \right)_\text{COL,kj} = \frac{\pi}{4} \bar{E}_{kj} N_k L_j F \left\{ \left( D_k (m_k) + D_j (m_j) \right)^2 \right\} G \left\{ \left[ v_k (\tilde{m}_k) - v_j (\tilde{m}_j) \right]^2 \right\}^{1/2} \tag{23}
\]

\[
\left( \frac{\partial N_k}{\partial t} \right)_\text{COL,kj} = - \frac{\pi}{4} \bar{E}_{kj} N_k N_j F \left\{ \left( D_k (\tilde{m}_k) + D_j (\tilde{m}_j) \right)^2 \right\} G \left\{ \left[ v_k (\tilde{m}_k) - v_j (\tilde{m}_j) \right]^2 \right\}^{1/2} \tag{24}
\]

\[
\left( \frac{\partial L_j}{\partial t} \right)_\text{COL,kj} = - \left( \frac{\partial L_k}{\partial t} \right)_\text{COL,kj}, \tag{25}
\]

with the functions \( F \) and \( G \) depending on the diameters \( D_k (\tilde{m}_k) \) and \( D_j (\tilde{m}_j) \), the terminal velocities \( v_k (\tilde{m}_k) \) and \( v_j (\tilde{m}_j) \), and the underlying size distribution of the different hydrometeors with mean masses \( \tilde{m}_k \) and \( \tilde{m}_j \) as discussed in Seifert and Beheng (2006). Thus, the rate of change in the mass density of a collector population [Eq. (23)] depends linearly on the collector number density and linearly on the available mass density of the collected particles—for example, for snow-cloud riming, Eq. (23) implies that the snow-cloud riming rate
increases with increasing snow crystal number density and increasing liquid water content. In the following, the collection of, say, cloud droplets by snow (ice, graupel) is also referred to as riming whereas the collection of, say, ice by snow (ice) is referred to as aggregation. For the collection of cloud droplets by ice and graupel, the mean collision efficiencies $E_{ic}$ and $E_{gc}$ are approximated by a piecewise linear function of the mean diameter (Seifert and Beheng 2006). For the collection of cloud droplets by snow (snow-cloud riming) to determine $E_{sc}$ we follow laboratory measurements by Lew et al. (1986) and approximate the mean collision efficiency as a function of the Stokes number $St$ such that

$$E_{sc} = 0.939St^{2.657}, \quad 0.01 \leq E_{sc} \leq 0.8,$$  \hspace{1cm} (26)

but we limit the mean collision efficiency within the range $E_{sc,min} = 0.01$ and $E_{sc,max} = 0.8$. The Stokes number $St$ is given by

$$St = \frac{2(u_s - u_c)u_c}{gD_s},$$  \hspace{1cm} (27)
determined by the mean terminal fall velocities of snow crystals $u_s$ and cloud droplets $u_c$, the maximum diameter of snowflakes $D_s$, and $g$, the acceleration due to gravity. The mean collision efficiency for snow-cloud riming $E_{sc}$ is shown in Fig. 1.

For fixed snow crystal size, the resulting collision efficiencies decrease with decreasing cloud droplet number size and also with increasing snow crystal size and fixed cloud droplet size. The sticking efficiencies (aggregation efficiencies) $E_{ks}$ for collisions between different ice particles $k$ with snow are parameterized depending on temperature after Lin et al. (1983) for snow, ice, and graupel and after Cotton et al. (1986) for the self-collection of ice:

$$E_{ks} = \exp[0.09(T - T_3)], \quad 0 \leq E_{ks} \leq 1,$$  \hspace{1cm} (28)

$$k \in \{i, s, g\},$$

$$E_{ii} = 10^{0.035(T - T_3) - 0.7}, \quad 0 \leq E_{ii} \leq 0.2.$$  \hspace{1cm} (29)

Here, $T_3 = 273.16$ K is the triple point of water. The sticking efficiencies increase with increasing temperature, which causes aggregation to be more efficient at warmer temperatures than at colder temperatures.

3. Model setup

a. Computational domain

The 3D computational domain is composed of 200 × 200 grid points in the horizontal with a grid spacing of 2 km, which yields a domain of $-200 \text{ km} \leq x \leq 200 \text{ km}$ along the $x$ axis and $-200 \text{ km} \leq y \leq 200 \text{ km}$ along the $y$ axis. A terrain-following smooth level vertical (SLEVE) coordinate system (Schär et al. 2002) is introduced in the vertical with 50 layers and a vertical grid spacing varying between 10 m in the lowermost and 1000 m in the uppermost layer. The model top is located at 21.5 km and the time step of the model is 20 s.

b. Idealized topography

The idealized topography has the form of a 3D mountain ridge (Kirshbaum and Durran 2005) such that

$$h(x, y) = \begin{cases} \frac{h_0}{16}(1 + \cos(\pi r))^4, & r \leq 1 \\ 0, & r > 1 \end{cases},$$  \hspace{1cm} (30)

and

$$r^2 = \begin{cases} \left(\frac{x - x_0}{4a}\right)^2 + \left(\frac{|y - y_0| - B}{4b}\right)^2, & |y - y_0| > B \\ \left(\frac{x - x_0}{4a}\right)^2, & |y - y_0| \leq B \end{cases}. $$  \hspace{1cm} (31)

Here, $h_0$ is the peak height of the mountain ridge, $a = 20 \text{ km}$ is the mountain half-width in the $x$ direction, and $b = 10 \text{ km}$ is the mountain half-width in the $y$ direction. The parameter $B = 30 \text{ km}$ controls the width of the ridge. The mountain range is centered in the computational domain at $x_0 = 100$ and $y_0 = 100$ in gridpoint space. Throughout this study, we use the idealized topography with a ridge height of $h_0 = 800 \text{ m}$.
c. Dynamical initialization

The model is initialized with a horizontally homogeneous basic state given by a dry atmosphere at rest with surface pressure $p_{SL}$ and surface temperature $T_{SL}$. The basic state is hydrostatically balanced and the temperature increases are constant with the logarithm of pressure such that $\frac{\delta T}{\delta \ln p} = 42 \text{ k} \ (\text{Dudhia} \ 1993; \text{Doms} \ \text{and Schättler} \ 2002)$. The initial horizontally homogeneous profiles of pressure $p(z)$ and temperature $T(z)$ are calculated analytically as a function of surface pressure $p_{SL}$, surface temperature $T_{SL}$, and the dry Brunt–Väisälä frequency $N_d$ (Clark and Farley 1984). In our simulations, the surface pressure is $p_{SL} = 1000 \text{ hPa}$ and the surface temperature is prescribed with temperatures $T_{SL} = 273 \text{ K}$ and $T_{SL} = 280 \text{ K}$, respectively. The dry Brunt–Väisälä frequency is chosen to be constant with height with $N_d = 0.011 \text{ s}^{-1}$. The vertical profile of the relative humidity is prescribed by a modified Fermi function of the type

$$\text{RH}(z) = a + \frac{b - a}{1 + \exp[-c(z - z_0)]}, \quad (32)$$

with the parameters $a = 0.95, b = 0.03, c = 0.0015 \text{ m}^{-1}, z_0 = 6000 \text{ m}$, and $0 \leq \text{RH} \leq 1$.

The horizontal wind profile $U$ is prescribed unidirectionally and the wind speed is vertically constant with $U = 15 \text{ m s}^{-1}$ up to 10 km and increases linearly above. The vertical profiles of temperature and dewpoint temperature are shown in the skew $T$–log $p$ diagram in Fig. 2. The cold sounding ($T_d = 273 \text{ K}$) is potentially and statically stable whereas the warm sounding ($T_d = 280 \text{ K}$) depicts an almost neutrally stratified atmosphere in the lower layers with marginal potential instability but increasing stability with height. The lifting condensation level (LCL) is located at approximately $z_{LCL} \approx 100 \text{ m}$ for all soundings whereas the freezing level varies. For the sounding $T_d = 273 \text{ K}$ the freezing level is located at the surface whereas for the sounding $T_d = 280 \text{ K}$ the freezing level is located at roughly 1000-m height.

Because the Rossby number $Ro = U/fL$ is usually much greater than unity for the characteristic scales considered here ($U = 15 \text{ m s}^{-1}, f = 10^{-4} \text{ s}^{-1}$ at middle-latitudes, and $L = 20 \text{ km}$), the effect of the Coriolis force is neglected in this study.

d. Microphysical initialization

To simulate realistic atmospheric aerosol conditions, the model is initialized with measurements obtained from the Cloud and Aerosol Characterization Experiment (CLACE; e.g., Choularton et al. 2008) held during February and March 2004 at the high-alpine research site Jungfraujoch (JFJ). The JFJ is usually influenced by remote-continental air masses but also by air masses with anthropogenic aerosol loads, which are entrained from the boundary layer because of slope wind circulations, convection, or frontal activity (Weingartner et al. 1999). Aerosol size distributions were determined with a scanning mobility particle sizer (SMPS) and an optical particle counter (OPC).

Throughout this study, the mean aerosol spectrum of March 2004 serves as the reference aerosol size distribution in the control simulations (CTL) and is shown in Fig. 3. The size distribution data from the SMPS and the OPC instruments are first averaged and then fitted with a trimodal exponential model (which is discussed in the appendix). The fitted parameters of the aerosol size distribution are summarized in Table 3. The aerosol mass distribution is calculated analytically from the aerosol number distribution by assuming spherical particles with a density of $\rho = 1.5 \text{ g cm}^{-3}$ (Cozic et al. 2007b).

We consider the reference aerosol size distribution to be mostly internally mixed and composed of sulfate, organic carbon, black carbon, and mineral dust. The assumption of the internal mixture corresponds to observations made at the JFJ for the fine-mode aerosols (PM 1) for most situations except, for example, Saharan dust events (Weingartner et al. 2002; Sjogren et al. 2008). For the coarse-mode aerosols we assume that roughly 30% of the aerosol number concentration is internally mixed with sulfate whereas the rest is assumed to be insoluble. Hence, the reference aerosol spectrum contains a few contact nuclei in the coarse mode. The estimate of approximately 30% mixed coarse-mode aerosols is deduced from the coarse-mode mass fraction, which was determined from an aerosol mass spectrometry (AMS) mass closure (Cozic et al. 2007b). For the mass fractionation of the different aerosol components we use the values from the CLACE 3 campaign given by Cozic et al. (2007b), which were obtained from an AMS mass closure. Sulfate nucleation-mode particles are assumed to form by homogeneous nucleation (gas to particle conversion) from the supersaturated gas-phase sulfuric acid concentration at low temperatures and, therefore, preferably in the upper troposphere. These nucleation-mode aerosols are smaller than 10 nm in diameter and are not available from the SMPS measurements. To get an estimate of the vertical distribution of the background gas-phase sulfuric acid concentration and the sulfate aerosols in nucleation mode, pseudosoundings are taken from the fifth version of the Max-Planck-Institut für Meteorologie GCM (ECHAM), ECHAM5-Hamburg Aerosol Model (HAM) model (Stier et al. 2005; Lohmann et al. 2007) for the nearest JFJ grid point as discussed in Muhlbauer and Lohmann (2008).
In the following we consider two distinct but typical events for the Alpine region that result in pronounced anomalies in the aerosol size distribution and especially in the aerosol composition. First, we consider a Saharan dust event that occurred during 13 and 14 March 2004 as a result of an intense anomaly in potential vorticity (PV) approaching North Africa and a strong advection of Mediterranean air masses toward the Alps in a south-westerly current (see Fig. 4). The aerosol anomaly for these days is shown in terms of a positive anomaly in the mean aerosol size distribution (see Fig. 3). The anomaly exhibits a pronounced deviation in the measured coarse-mode aerosol number concentrations of the OPC. The count median radius of the dust anomaly is roughly 1.3 μm. Ensemble backward trajectory calculations with the Lagrangian analysis tool (LAGRANTO; Wernli and Davies 1997; Wernli 1997) based on 6-hourly European Centre for Medium-Range Weather Forecasts (ECMWF) analysis fields with a resolution of 0.3° are shown in Fig. 5. Although the exact origin of the air mass is unclear, all trajectories start in the North African boundary layer and thus support the assumption of mineral dust in the aerosol anomaly.

The second aerosol anomaly is from 8 March 2004 when Switzerland was influenced by continental air masses with anthropogenic contributions. The anomaly shows a pronounced deviation in the Aitken mode at approximately 0.02 μm and in the accumulation mode.
at roughly 0.07 μm. Measurements by Cozic et al. (2007b) support the assumption of anthropogenic aerosols because a relatively high fraction of the fine-mode aerosol mass was found to be black carbon. Backward trajectory calculations corroborate the assumption of continental aerosols but show two distinct air mass contributions. One contribution is from an air mass approaching Switzerland from a northwesterly direction and the second contribution originates from the southeast. The synoptic-scale flow conditions shown in Fig. 4 indicate that the northwesterly branch is connected to the cold-air advection behind the upper-level trough whereas the southeasterly branch is connected to southeasterly flows at lower levels and a frontal system impinging on the Alps. Both air masses originate from

![Image: Figure 3. Mean number and mass distribution of aerosols measured at the JFJ during the CLACE 3 field campaign in March 2004. (a) The mean number distribution of aerosols measured with an SMPS (triangles) and an OPC (squares) together with a trimodal exponential model (solid) fitted to the data. The contributions of the Aitken (dashed), accumulation (dashed–dotted), and coarse mode (dotted) are indicated. (b) The mean mass distribution of aerosols (solid) calculated analytically from the number distribution by assuming spherical particles with a density of ρ = 1.5 g cm⁻³. (c), (d) The positive anomalies of the aerosol number distribution with respect to the mean aerosol spectrum during CLACE 3 for (c) 13 and 14 Mar 2004 and (d) 8 Mar 2004.

| TABLE 3. Parameters of the aerosol size distributions. The reference aerosol spectrum serves as the control (CTL) and the perturbations are the mineral dust anomaly (DUA) and the fine-mode anomaly (BCA). |
|---|---|---|---|---|---|
| Mode | N (cm⁻³) | r (μm) | σ | M (μg m⁻³) | Measuring device |
| CTL AIT | 723 | 0.014 | 2.28 | 0.28 | SMPS |
| CTL ACC | 221 | 0.067 | 1.66 | 1.34 | SMPS |
| CTL COA | 0.43 | 0.707 | 1.57 | 2.37 | OPC |
| DUA COA | 1.34 | 1.316 | 1.56 | 46.0 | OPC |
| BCA AIT | 359 | 0.019 | 1.36 | 0.02 | SMPS |
| BCA ACC | 225 | 0.068 | 1.58 | 1.12 | SMPS |
lower atmospheric layers but the southeasterly branch undergoes a very intense lifting as the frontal system impinges on the main Alpine rim.

In the following experiments, we consider these different aerosol anomalies as typical and realistic aerosol perturbations for the Alpine region and add the perturbation aerosol size distribution on top of the reference aerosol spectrum. The natural dust anomaly is assumed to be insoluble with only a small fraction of dust internally mixed with sulfate, which yields an aerosol perturbation with mainly additional contact freezing nuclei and only a few immersion and cloud condensation nuclei. In the following section the simulations with the dust anomaly are denoted as DUA. The anthropogenic aerosol anomaly is assumed to consist of internally mixed black carbon for the accumulation mode and freshly formed insoluble black carbon in the Aitken mode. Thus, the anthropogenic aerosol anomaly adds additional cloud condensation nuclei as well as contact and immersion nuclei to the reference aerosol spectrum. The simulation with the black carbon anomaly is denoted as BCA.

4. Experiment A: Simulations with dust

The following experiments use the initial conditions discussed in section 3 and compare the control simulation with the simulation including enhanced dust aerosols.
The nondimensional mountain height \( \hat{h} = Nh/U \) for this setting is approximately 0.6, suggesting the development of a linear hydrostatic mountain wave (Smith 1979, 1980). The orographic flow is supposed to pass predominantly over instead of around the mountain ridge.

As a result of the forced upslope ascent, an orographic cloud forms at the windward side of the mountain range (shown in Fig. 6 for the control simulation).

The cloud base is located at the lifting condensation level at approximately \( z_{LCL} \approx 100 \) m and the vertical extent of the cloud is roughly 3000 m. The orographic cloud indicates a slight upstream tilt, which is directly connected with the phase tilt of the mountain wave. The low-level cloud consists mainly of supercooled liquid water but also shows contributions of cloud ice formed by contact freezing. Additionally, an orographic wave cloud that consists solely of ice induced by immersion freezing forms at upper levels. The collection of cloud ice by liquid water droplets leads to the growth of graupel particles, which immediately precipitate. Snow forms by aggregation of ice crystals and is transported far into the leeward side of the mountain because of the small terminal fall velocities. The contribution of rain is negligibly small and is not shown in Fig. 6.

The precipitation field (Fig. 7) shows contributions of snow and graupel close to the mountain top as well as

![Fig. 5](image-url)
contributions from snow far downstream of the mountain range. The latter is caused by precipitation from an upper-level wave cloud but the contribution in terms of total precipitation is generally low for these types of clouds (Jiang and Smith 2003).

The simulation with enhanced dust aerosols shows a similar cloud structure but the ice phase at lower levels is more pronounced and occurs already on the upstream side of the mountain (Fig. 8).

The increase in the ice water content is caused by an increase in the freezing rates induced by the additional contact nuclei; as a consequence, the liquid water content in the orographic cloud is reduced. The enhanced initiation of cloud ice by contact freezing enhances the precipitation efficiency of the low-level orographic cloud, which leads to an increase in graupel and snow and thus to a precipitation enhancement on the upstream side of the mountain and close to the mountain peak.

The precipitation differences shown in Fig. 9 reveal an increase in upslope precipitation and a slight reduction of downslope precipitation caused by the earlier onset of the ice phase in the DUA simulation. The increased upslope precipitation also leads to stronger downdrafts on the lee side of the mountain range, which in turn lead to an increase in surface temperature and to a raising of the melting level on the leeward side. The raise in the leeward melting level explains the slight reduction of snow and the increase in rain far downstream of the mountain.

Figure 10 shows the volume mean mass conversion rates for the most important processes leading to
precipitation, which are aggregation, riming, and warm-phase collection. In the subsequent context we refer to warm-phase collection as the sum of cloud droplet conversion to rain (autoconversion) and accretion. Riming includes the collection of cloud water by ice crystals and snowflakes as well as the possible conversion to graupel. Aggregation includes the collection of ice and snow.

For both simulations (CTL and DUA) at cold temperatures ($T_{sl} = 273$ K; Fig. 10, left) the dominant processes are aggregation followed by riming whereas the contribution of warm-phase collection is almost negligible. An increase in the number concentration of dust aerosols leads to an increase in the available contact nuclei and to an increase of the ice phase induced by contact freezing. The increased glaciation of the cloud reduces the liquid water content, which also reduces the warm-phase collection rates such as autoconversion and accretion. The riming and aggregation rates are both increased in the simulation DUA, but the increase is more pronounced for riming than for aggregation. Because riming and aggregation are the dominant processes at cold temperatures, the increase in the conversion rates of riming and aggregation leads to an increase of the total mass conversion rates and thus to an increase in precipitation.

For warmer temperatures ($T_{sl} = 280$ K; Fig. 10, right) the dominant microphysical processes in the control simulation are again aggregation followed by riming and warm-phase collection. Generally, aggregation becomes more important if the temperature increases because the sticking efficiency increases severely with increasing temperature. In contrast to the simulations with cold temperatures, collection (especially accretion) becomes more important at warmer temperatures. Enhancing the dust aerosol number concentration...
leads to an increase in riming and aggregation and, again, enhances orographic precipitation. Furthermore, the warm-phase collection rates are increased in the simulation DUA because of the melting of snowflakes to rain, which subsequently increases the accretion rates.

5. Experiment B: Simulations with black carbon

In the next experiment we change the aerosol-microphysical initialization while keeping the dynamical initial condition constant. The aerosol perturbation is considered to be anthropogenic black carbon (BCA) in order to investigate the potential role of the anthropogenic perturbations on the orographic precipitation. Similar to the simulations with dust, the amount of upslope precipitation is increased for cold as well as for warm initial surface temperatures (shown in Figs. 11 and 12). The precipitation is falling in form of snow and graupel in the cold simulation and in form of graupel and rain in the warm simulation. For the cold simulations the most dominant cloud-microphysical processes leading to precipitation are, again, aggregation and riming, with an almost negligible contribution of warm-phase collection shown in Fig. 13. Introducing additional contact and immersion nuclei leads to an overall increase of the riming and aggregation rates but to an overall decrease in the warm-phase collection due to the additionally available cloud condensation nuclei. Similar results are obtained if the surface temperature is changed from $T_{sl} = 273$ K to $T_{sl} = 280$ K, but here the warm-phase collection is increased with increasing CCN due to the melting of snow and the subsequent increase of accretion.
Because the availability of contact nuclei in the initial aerosol perturbation field is uncertain we test the sensitivity regarding the mixing state of the aerosol spectrum. Previously we assumed that only the accumulation-mode aerosols are internally mixed and the Aitken-mode aerosols are freshly formed black carbon aerosols and serve as contact nuclei. Next, we test the sensitivity in the orographic precipitation pattern if a fully internally mixed aerosol perturbation spectrum is considered. As a consequence, only cloud condensation and immersion nuclei are added to the reference aerosol spectrum. The immersion nuclei require lower temperatures for inducing freezing than do contact nuclei according to the measurements summarized in Diehl and Wurzler (2004). The simulations with the fully internally mixed aerosol perturbation are denoted as BCM. Figure 12 shows that in this case the perturbed simulation gives slightly lower total domain precipitation than the control simulation for the cold simulation and an almost equal precipitation in the warm simulation. In contrast to the previous simulation, the additionally available CCN lead to a decrease in the warm-phase collection rates (autoconversion and accretion) and to a decrease in riming in both (cold and warm) simulations. However, aggregation is only marginally affected. Because aggregation is the dominant cloud-microphysical process in the simulations, the net effect is a slight decrease in the total mass conversion rates and thus a slight decrease in orographic precipitation. The simulations with warmer temperature \((T_{SL} = 280 \text{ K})\) show almost no change in precipitation.

**FIG. 9.** Precipitation difference at the center line for experiment A after 10 h. Shown is the precipitation difference (DUA − CTL) of rain (blue), snow (red), graupel (green), and total precipitation (black) for the simulations (a) \(T_{sl} = 273 \text{ K}\) and (b) \(T_{sl} = 280 \text{ K}\), respectively.

**FIG. 10.** Mass conversion rates \((\text{kg} \text{ m}^{-3} \text{ s}^{-1}, \text{solid})\) and the contributing conversion rates of warm-phase (wp) collection (dotted), aggregation (dashed–dotted), and riming (dashed) for experiment A for \(T_{sl} = (a) 273 \text{ and } (b) 280 \text{ K}\).
6. Discussion and conclusions

Idealized 3D simulations of orographic precipitation from a linear hydrostatic mountain wave are conducted. The simulations are microphysically initialized with aerosol perturbations that are typical and representative for the Alpine region and central Switzerland. Two different types of aerosols—naturally occurring mineral
dust and anthropogenic black carbon—are considered. The potential role of these distinct types of aerosols in influencing microphysical processes in orographic clouds and precipitation is investigated at different temperatures. A summary of the experiments is given in Table 4.

Because mineral dust particles are efficient ice nuclei in the contact mode, the enhancement of dust in the ambient air leads to an early initiation of the ice phase in orographic clouds and to more pronounced ice water content on the upslope side of the mountain range. The increased glaciation of the orographic cloud by contact freezing increases riming and aggregation but decreases the warm-phase collection. Because aggregation and riming are the dominant processes in the simulations with a low freezing level, the early initiation of the ice phase in orographic clouds enhances the efficiency of these microphysical processes. The net effect leads to an increase in the precipitation efficiency, which in turn increases orographic precipitation due to an increase in snow and graupel. Increasing the ambient temperature changes the relative importance of aggregation, riming, and warm-phase collection in the orographic cloud but does not change the net effect of increased orographic precipitation.

The model simulations suggest that anthropogenic aerosols that are capable of acting as contact ice nuclei can increase orographic precipitation similarly to naturally occurring contact ice nuclei such as mineral dust. Although the freezing efficiency of black carbon in the contact mode is lower than the freezing efficiency of mineral dust, the overall increase in the orographic precipitation efficiency may be larger in the case of black carbon than in the case of mineral dust. The explanation for this effect is twofold. First, the collision efficiency of aerosols and cloud droplets induced by Brownian diffusion is larger for small black carbon aerosols than for large dust aerosols because the aerosol–cloud droplet
collision efficiency is inversely related to the aerosol size. This leads to a more pronounced glaciation in the case of black carbon even if the material-specific freezing efficiency is lower for black carbon than for mineral dust. Second, the typically occurring perturbations in terms of the aerosol number density are much lower in the case of mineral dust than in the case of black carbon. For example, typical values for the anomaly in the aerosol number density are on the order of 0.1 to 1 cm$^{-3}$ in the case of mineral dust whereas anomalies are on the order of 100 to 1000 cm$^{-3}$ in the case of anthropogenically induced black carbon. Thus, the large collision efficiency together with the large amount of available contact nuclei may render anthropogenically induced black carbon aerosols highly efficient at glaciating orographic clouds and enhancing orographic precipitation. However, uncertainties remain with respect to the freezing efficiency of small black carbon aerosols and more measurements are needed.

Our simulations suggest that the mixing state of the aerosol anomaly plays a crucial role because coating or mixing may deactivate potential ice nuclei in the contact mode (Storelvmo et al. 2008; Hoose et al. 2008). The latter are important for initiating the ice phase in orographic clouds at warmer temperatures and thus at lower levels and earlier in the cloud. As a consequence

![Table 4](image)

**Table 4.** Summary of mean mass conversion rates by riming, aggregation, and collection for the different experiments. The +/− sign indicates if the mass conversions due to riming, aggregation, or warm-phase collection are increased or decreased whereas ~ denotes a change on the order of 1%. The number in parentheses denotes the dominance of the microphysical process in terms of the total conversion rates in descending order.

<table>
<thead>
<tr>
<th>Simulation</th>
<th>$T_{sl}$ (K)</th>
<th>Riming</th>
<th>Aggregation</th>
<th>WP collection</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>DUA</td>
<td>273</td>
<td>+, (2)</td>
<td>+, (1)</td>
<td>−, (3)</td>
<td>−</td>
</tr>
<tr>
<td>DUA</td>
<td>280</td>
<td>+, (2)</td>
<td>+, (1)</td>
<td>+, (3)</td>
<td>+</td>
</tr>
<tr>
<td>BCA</td>
<td>273</td>
<td>+, (2)</td>
<td>+, (1)</td>
<td>−, (3)</td>
<td>+</td>
</tr>
<tr>
<td>BCA</td>
<td>280</td>
<td>+, (2)</td>
<td>+, (1)</td>
<td>+, (3)</td>
<td>+</td>
</tr>
<tr>
<td>BCM</td>
<td>273</td>
<td>−, (2)</td>
<td>−, (1)</td>
<td>−, (3)</td>
<td>−</td>
</tr>
<tr>
<td>BCM</td>
<td>280</td>
<td>−, (3)</td>
<td>~, (1)</td>
<td>−, (2)</td>
<td>−</td>
</tr>
</tbody>
</table>
of coating and/or mixing, potential ice nuclei in the contact mode may be deactivated and may serve subsequently only in the immersion mode and thus at lower temperatures.

Our simulations suggest that the proposed mechanism of decreased riming rates with increasing aerosol load is reproducible in a numerical model only if a fully internally mixed aerosol spectrum is assumed. In this case, the aerosol anomaly does not provide any further contact nuclei, which are important for an early formation of the ice phase in orographic clouds. Furthermore, the increase in CCN becomes dominant, which leads then to a decrease in riming and warm-phase collection and to a decrease of the orographic precipitation.

Based on our model simulations, heterogeneous ice nucleation induced by aerosols appears to be an important ingredient for the problem of aerosol–cloud interactions, the initiation of ice in mixed-phase orographic clouds, and the possible implications for orographic precipitation. However, further measurements of aerosols and cloud-microphysical variables are needed to narrow down the current uncertainty in heterogeneous ice nucleation processes.

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APPENDIX

Fitting Aerosol Size Distributions

The observed aerosol number density distribution is fitted using an exponential model of the form

$$ f(\ln r) = \sum_{k=1}^{K} \hat{a}_k \exp \left[ - \left( \frac{\ln r - \hat{b}_k}{\hat{c}_k} \right)^2 \right], \tag{A1} $$

with $K = 1$ for a monomodal distribution, $K = 2$ for a bimodal distribution, and $K = 3$ for a trimodal distribution. The three times $K$ coefficients $\hat{a}_k$, $\hat{b}_k$, and $\hat{c}_k$ are determined with robust nonlinear least squares regression. The relations among the estimated number densities $\hat{N}_k$, the count median radii $\hat{r}_k$, and the geometric standard deviations $\hat{\sigma}_k$ are determined by mapping the coefficients from the fitted exponential model to a log-normal distribution that involves the following transformations for the coefficients:

$$ \hat{N}_k = \sqrt{\pi} \hat{a}_k \hat{c}_k, \tag{A2} $$

$$ \hat{r}_k = \exp(\hat{b}_k), \tag{A3} $$

$$ \hat{\sigma}_k = \exp \left( \frac{\hat{c}_k}{\sqrt{2}} \right). \tag{A4} $$

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