A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles

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ABSTRACT
In climate and weather models, the quantitative description of aerosol and cloud processes relies on simplified assumptions. This contributes major uncertainties to the prediction of global and regional climate change. Therefore, models need good parameterizations for heterogeneous ice nucleation by atmospheric aerosols. Here the authors present a new parameterization of immersion freezing on desert dust particles derived from a large number of experiments carried out at the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) cloud chamber facility. The parameterization is valid in the temperature range between −12°C and −36°C at or above water saturation and can be used in atmospheric models that include information about the dust surface area. The new parameterization was applied to calculate distribution maps of ice nuclei during a Saharan dust event based on model results from the regional-scale model Consortium for Small-Scale Modelling–Aerosols and Reactive Trace Gases (COSMO-ART). The results were then compared to measurements at the Taunus Observatory on Mount Kleiner Feldberg, Germany, and to three other parameterizations applied to the dust outbreak. The aerosol number concentration and surface area from the COSMO-ART model simulation were taken as input to different parameterizations. Although the surface area from the model agreed well with aerosol measurements during the dust event at Kleiner Feldberg, the ice nuclei (IN) number concentration calculated from the new surface-area-based parameterization was about a factor of 13 less than IN measurements during the same event. Systematic differences of more than a factor of 10 in the IN number concentration were also found among the different parameterizations. Uncertainties in the modeled and measured parameters probably both contribute to this discrepancy and should be addressed in future studies.

1. Introduction
As one of the major components of the tropospheric aerosol, desert dust particles influence the climate system both directly by scattering and absorbing shortwave solar radiation (e.g., Tegen 2003) and indirectly by influencing...
the formation and life cycles of tropospheric clouds (e.g., Levin et al. 1996; Rosenfeld et al. 2001; Kelly et al. 2007; Hoose et al. 2008). Large desert areas in northern Africa (Sahara, Bodélé Depression) and Asia (Taklimakan, Gobi) are known as major source areas of atmospheric desert dust particles (Prospero et al. 2002; Tegen 2003).

Although desert dust is emitted in extremely dry air, a trajectory analysis by Wiedeck et al. (2010) demonstrated that a significant fraction of dusty air masses originating from African and Asian desert areas experience cloud formation conditions after some time of transport and lifting in the troposphere. In situ measurements and lidar studies have demonstrated a link between long-range transported Saharan dust and ice formation in tropospheric clouds (DeMott et al. 2003; Sassen et al. 2003; Ansmann et al. 2008).

Ice formation by aerosol particles can occur by homogeneous or heterogeneous nucleation (Pruppacher and Klett 1997). Homogeneous freezing of water droplets denotes the spontaneous formation of ice from a supercooled liquid droplet in the absence of any foreign solid phase at temperatures below −36°C. Heterogeneous ice nucleation takes place in the presence of ice nuclei (IN), a minor fraction of the tropospheric aerosol, which exhibit specific surface structures to facilitate the formation of ice crystals. With the presence of IN, clouds below 0°C, which are primarily composed of supercooled liquid droplets, can start to develop the ice phase. Heterogeneous ice nucleation can occur either through the direct deposition of water from the vapor phase to the dry surface of solid aerosol particles (deposition ice nucleation mode) or by the freezing of a supercooled liquid phase in contact with the ice nucleus (freezing modes). In the latter case, the ice nucleus can already be immersed in the supercooled liquid when freezing happens (immersion freezing mode). It was first shown in Young (1974), who discussed surface densities of active sites in relation to contact freezing and deposition nucleation.

Based on new series of ice nucleation studies using the Aerosol Interactions and Dynamics in the Atmosphere (AIDA) cloud chamber facility at the Karlsruhe Institute of Technology (KIT), a straightforward approach was applied to calculate the IN number concentration as a function of temperature and dust particle surface area. This parameterization was used to calculate distribution maps of immersion freezing nuclei during a Saharan dust event based on model results from the regional-scale online coupled model system Consortium for Small-Scale Modelling–Aerosols and Reactive Trace Gases (COSMO-ART; Vogel et al. 2009; Bangert et al. 2011a). This was then compared to measurements at the Taunus Observatory on Mount Kleiner Feldberg (located 25 km northwest of Frankfurt am Main, Germany, at 825-m altitude), and to other ice nucleation parameterizations, including the parameterizations by DeMott et al. (2010), Hoose et al. (2010), and Phillips et al. (2008), during a Saharan dust outbreak event in May 2008 (Klein et al. 2010).

In section 2 we describe the experimental setup and dust samples used in this study. In section 3 a typical immersion freezing experiment is described, followed by the results of this study (section 4). In section 4b we derive the new parameterization of immersion freezing on desert dust particles. In section 5a the model COSMO-ART is described. Section 5b presents the application of the new parameterization, followed by a description of the measurements at the Taunus Observatory in section 5c. In section 5d we compare the calculated IN number concentration to the measurements and to other concepts (section 5e). The results are summarized in section 6.
2. Experimental setup

a. The AIDA cloud simulation chamber

The cloud simulation chamber AIDA (Möhler et al. 2006) of KIT was used to investigate immersion freezing on desert dust particles. The central part of the AIDA facility (Fig. 1) is a large, evacuable vessel with a volume of 84 m³, which can be used as a cloud expansion chamber for ice nucleation studies. It allows a well-controlled and repeatable simulation of cooling rates in the range of 0.1–6.0 K min⁻¹, which corresponds to updraft velocities of about 0.15–8 m s⁻¹ in natural wave clouds or convective clouds. The cylindrical-shaped aluminum chamber is encased in a large thermally insulated box that can be cooled down to a minimum temperature of −90°C, with a homogeneous control of constant temperatures within ±0.3 K throughout the box. The cloud chamber itself is connected to a vacuum and synthetic air supply system. It can be evacuated to less than 0.01 hPa. It is flushed and filled with clean synthetic air prior to each experiment. After cleaning and filling, the particle background in the chamber is typically less than 0.1 cm⁻³. At constant pressure and temperature prior to a cloud expansion experiment, almost ice saturated conditions in the cloud chamber (typically about 90%–95% relative humidity with respect to ice) are maintained by a thin ice layer on the chamber walls. A mixing fan is installed near the chamber bottom to ensure homogeneous conditions during the experiments. At constant temperature and pressure control, the spatial and temporal temperature fluctuations throughout the cloud chamber are less than ±0.2 K.

b. Instrumentation

Figure 1 shows a schematic of the cloud chamber and the instrumentation used in this study. A detailed description can be found in, for example, Möhler et al. (2006) and Wagner et al. (2006).

The desert dust samples described in section 2c were introduced into the chamber using the rotating brush disperser RBG-1000 (Palas GmbH, Karlsruhe, Germany). The total particle number concentration was measured with a condensation particle counter (CPC3010; TSI Inc., Shoreview, MN). A Scanning Mobility Particle Sizer (SMPS; TSI Inc.) and an Aerodynamic Particle Sizer (APS, TSI Inc.) measured the number size distribution before each experiment. Measured size distributions are presented and will be discussed in section 4a.

A tunable diode laser (TDL) absorption spectrometer was applied for the in situ measurement of the water vapor pressure with an accuracy of about 5% (Ebert et al. 2005; Fahey et al. 2009). The saturation vapor pressure formulations of Murphy and Koop (2005) were used to calculate the relative humidity with respect to ice and water using the AIDA mean gas temperature and the measured water vapor pressure. Additionally, the total water concentration was measured with a chilled-mirror hygrometer. This accurate absolute measurement for total water was compared to the TDL absorption spectrometer in cloud-free situations, which yielded an agreement between both instruments within about 1%–2%.

Two Welas optical particle counters (Palas GmbH) were used to detect and count cloud droplets and ice crystals (Benz et al. 2005). The detection ranges of the two instruments are 0.3–46 μm (Welas1) and 4–237 μm (Welas2), respectively. Ice crystals were distinguished from droplets and interstitial aerosol by their optical size. Since the saturation vapor pressure over liquid is greater than over ice at the same temperature, liquid water droplets evaporate in the presence of ice crystals. The ice crystals grow quickly at the expense of the liquid water droplets (Wegener–Bergeron–Findeisen process). For example, the growth rate of a 10-μm ice crystal at liquid water saturation is about 3.4 μm s⁻¹ at −15°C. In the analysis, a size threshold, which varied among the experiments, was set so that the number of all particles with sizes above the threshold could be used to calculate the number concentration of ice crystals. For this calculation we used the Welas2 data only. The reason for this is the higher sensitivity of the Welas2 instrument compared to Welas1. The ice number concentration was calculated for every 5 s and then averaged over a 60-s time period, making sure that at least four ice crystals were counted in this time interval. Otherwise, the time interval was increased until this condition was satisfied, leading to a higher uncertainty in the corresponding temperature. The averaging started at droplet formation.
c. Dust samples

Five dust samples were examined in this study: Asian dust (AD), Saharan dust (SD), Canary Island dust (CID), Israel dust (ID), and Arizona test dust (ATD). The origins of the dust samples are listed in Table 1. AD, SD, CID, and ID were collected from the surface. They are samples of natural origin and are considered to be representative for atmospheric dust. ATD (Powder Technology Inc., Burnsville, MN) is a commercially available reference dust material. Here we used an ATD sample with a nominal 0–3-μm particle diameter size range [see Möhler et al. (2006) for further details].

The AD, SD, CID, and ID samples were sieved, but not chemically or otherwise processed, to obtain a subfraction with particle diameters smaller than 75 μm that were used for aerosol generation. The dust samples were introduced into the chamber using the rotating brush disperser, which was operated with dry and clean synthetic air to avoid contamination. Before adding the aerosol to the cloud chamber, large dust particles were removed with a cyclone impactor operated with a 50% cutoff between about 1 and 5 μm. For a few experiments as indicated in Table 2 no cyclone was used in order to generate a dust aerosol with larger particles for the cloud experiments. In this case particles larger than about 10 μm were still removed by settling and inertial losses of particles in the tube connection between the dust generator and the cloud chamber.

3. AIDA expansion experiments

The immersion freezing on desert dust particles was investigated during the International Workshop on Comparing Ice Nucleation Measuring Systems (ICIS07) in September 2007 (Nicolet et al. 2010; Koehler et al. 2010; Kanji et al. 2011; Jones et al. 2011) and during the Aerosol–Cloud Interaction campaign ACI04 in September/October 2010. An overview of the experiments is given in Table 2.

Each experiment started with the evacuation of the chamber volume to less than 0.1 hPa and refilling it with clean synthetic air to ensure clean conditions. Also a reference expansion was performed before each experiment, where the background aerosol was activated to droplet freezing.

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**Table 1. Dust types and origin of samples.**

<table>
<thead>
<tr>
<th>Dust</th>
<th>Origin</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD</td>
<td>Soil sample collected in the eastern parts of the Takla Makan Desert in China; provided by Lothar Schütz, University of Mainz, Germany</td>
</tr>
<tr>
<td>SD</td>
<td>Soil sample collected about 50 km north of Cairo, Egypt</td>
</tr>
<tr>
<td>CID</td>
<td>Soil sample collected near the town of Mala on the Canary Island of Lanzarote</td>
</tr>
<tr>
<td>ID</td>
<td>Collected as sedimented particles after a dust storm in Israel; provided by Prof. Eli Ganor, Tel Aviv University</td>
</tr>
<tr>
<td>ATD</td>
<td>Milled reference dust material, Powder Technology, Inc., Minnesota</td>
</tr>
</tbody>
</table>

**Table 2. Overview of immersion freezing experiments and respective experimental conditions.**

<table>
<thead>
<tr>
<th>Expt No.</th>
<th>Date</th>
<th>Dust</th>
<th>$p_0$ (hPa)</th>
<th>$T_0$ (K)</th>
<th>$T_{range}$ (K)</th>
<th>$N_{tot}$ (cm$^{-3}$)</th>
<th>$S_{tot}$ (μm$^2$ cm$^{-3}$)</th>
<th>Cyclone</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>ACI04_13</td>
<td>1 Oct 2010</td>
<td>AD</td>
<td>1003.3</td>
<td>−10.0</td>
<td>−8.0</td>
<td>173.3</td>
<td>109.2</td>
<td>Yes</td>
<td>No ice</td>
</tr>
<tr>
<td>ACI04_16</td>
<td>1 Oct 2010</td>
<td>AD</td>
<td>1002.8</td>
<td>−14.3</td>
<td>−8.1</td>
<td>238.3</td>
<td>130.0</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_19</td>
<td>4 Oct 2010</td>
<td>AD</td>
<td>990.1</td>
<td>−20.1</td>
<td>−8.0</td>
<td>218.2</td>
<td>156.4</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_04</td>
<td>29 Sep 2010</td>
<td>SD</td>
<td>1007.5</td>
<td>−9.1</td>
<td>−7.9</td>
<td>127.8</td>
<td>145.9</td>
<td>Yes</td>
<td>No ice</td>
</tr>
<tr>
<td>ACI04_37</td>
<td>7 Oct 2010</td>
<td>SD</td>
<td>1008.2</td>
<td>−9.8</td>
<td>−5.6</td>
<td>114.8</td>
<td>248.6</td>
<td>No</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_07</td>
<td>30 Sep 2010</td>
<td>SD</td>
<td>1005.5</td>
<td>−14.1</td>
<td>−8.0</td>
<td>173.9</td>
<td>189.2</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
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<td>ACI04_34</td>
<td>6 Oct 2010</td>
<td>SD</td>
<td>999.4</td>
<td>−14.8</td>
<td>−7.7</td>
<td>138.2</td>
<td>309.2</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ICIS07_20</td>
<td>25 Sep 2007</td>
<td>SD</td>
<td>1005.7</td>
<td>−16.4</td>
<td>−4.4</td>
<td>396.8</td>
<td>98.5</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_10</td>
<td>30 Sep 2010</td>
<td>SD</td>
<td>1003.9</td>
<td>−19.3</td>
<td>−8.2</td>
<td>121.8</td>
<td>110.2</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ICIS07_23</td>
<td>26 Sep 2007</td>
<td>CID</td>
<td>1002.0</td>
<td>−18.4</td>
<td>−5.1</td>
<td>381.8</td>
<td>59.1</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_25</td>
<td>5 Oct 2010</td>
<td>ID</td>
<td>995.7</td>
<td>−9.8</td>
<td>−7.5</td>
<td>695.6</td>
<td>576.7</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_40</td>
<td>7 Oct 2010</td>
<td>ID</td>
<td>1007.9</td>
<td>−9.4</td>
<td>−7.9</td>
<td>739.5</td>
<td>1229.7</td>
<td>No</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_28</td>
<td>5 Oct 2010</td>
<td>ID</td>
<td>996.4</td>
<td>−14.4</td>
<td>−6.6</td>
<td>655.4</td>
<td>514.6</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_31</td>
<td>6 Oct 2010</td>
<td>ID</td>
<td>1000.9</td>
<td>−15.0</td>
<td>−7.8</td>
<td>766.5</td>
<td>1333.9</td>
<td>No</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_43</td>
<td>8 Oct 2010</td>
<td>ID</td>
<td>1007.1</td>
<td>−15.0</td>
<td>−8.3</td>
<td>526.6</td>
<td>922.5</td>
<td>No</td>
<td>Freezing</td>
</tr>
<tr>
<td>ICIS07_17</td>
<td>24 Sep 2007</td>
<td>ID</td>
<td>1005.1</td>
<td>−16.8</td>
<td>−6.6</td>
<td>253.8</td>
<td>110.3</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ACI04_22</td>
<td>4 Oct 2010</td>
<td>ID</td>
<td>989.2</td>
<td>−19.6</td>
<td>−7.9</td>
<td>252.1</td>
<td>165.5</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
<tr>
<td>ICIS07_03</td>
<td>17 Sep 2007</td>
<td>ATD</td>
<td>1000.9</td>
<td>−16.8</td>
<td>−4.9</td>
<td>950.0</td>
<td>328.0</td>
<td>Yes</td>
<td>Freezing</td>
</tr>
</tbody>
</table>
or ice crystals and precipitated from the cloud chamber. Then a fresh sample of dust was dispersed into the cloud chamber. Figure 2 shows a typical AIDA immersion freezing experiment (experiment ACI04_22) with ID starting at an initial temperature of $219.6^\circ C$ with atmospheric pressure (Fig. 2, row 1). Comparable time series were obtained for all other experiments listed in Table 2.

The pumping expansion starts at $t = 0$ s. The gas temperature $T_{\text{gas}}$ starts to decrease almost adiabatically, while the wall temperature $T_{\text{wall}}$ stays nearly constant (Fig. 2, row 1). Because of the heat flux from the warmer chamber walls, $T_{\text{gas}}$ deviates from the adiabatic profile as $T_{\text{wall}} - T_{\text{gas}}$ increases (not shown).

The experiment starts at nearly ice saturated conditions (RH$_{\text{ice}}$ in Fig. 2, row 2). The initial relative humidity with respect to water (RH$_{\text{water}}$) is 80.5%. Because of the cooling expansion RH$_{\text{ice}}$ exceeds 100% after a few seconds. Water saturated conditions (RH$_{\text{water}} = 100\%$) are reached after about 75 s. Note that RH$_{\text{water}}$ had to be corrected by 5% in order to match water saturated conditions during the presence of water droplets in the cloud chamber. This systematic deviation can be caused by a slight offset in the temperature measurement due to the condensation of water vapor on the temperature sensors or by inhomogeneities especially close to the chamber walls that lead to lower calculated average humidity compared with the regions where cloud droplets are present.

The total particle number concentration is shown in row 3 of Fig. 2 (aerosol total). It is calculated from the initial CPC3010 value measured before the expansion start multiplied by the pressure dilution $p/p_0$, assuming a constant particle mixing ratio in the course of the experiment. The direct CPC3010 measurement is not shown here because it is affected by particle losses in the unheated sampling tube after droplet activation. Droplets and ice crystals are measured by the two optical particle counters Welas1 and Welas2. The total particle number concentration measured by Welas1 (Welas1 all in Fig. 2, row 3) increases by the formation of liquid droplets as soon as RH$_{\text{water}}$ reaches the cloud condensation nucleation threshold for the dust particles. After the cloud formation, Welas1 all equals the total number concentration of cloud droplets at about 75 s. (row 4) Particle size distribution with size threshold between droplets and ice crystals at $d_\rho = 30 \mu m$. See section 3 for more details.

![Diagram](https://via.placeholder.com/150)

**Fig. 2.** AIDA cloud expansion experiment (experiment ACI04_22) with Israel dust. Initial temperature is $-19.6^\circ C$. (row 1) Pressure $p$, wall temperature $T_{\text{wall}}$, gas temperature $T_{\text{gas}}$; (row 2) relative humidity with respect to water (RH$_{\text{water}}$) and ice (RH$_{\text{ice}}$); and (row 3) number concentration of total aerosol and ice crystals (Welas1 ice, Welas2 ice). Welas1 all equals the total number concentration of cloud droplets after cloud formation at about 75 s. (row 4) Particle size distribution with size threshold between droplets and ice crystals at $d_\rho = 30 \mu m$. See section 3 for more details.
crystals when they grow to an optical diameter larger than a certain size threshold, which is set individually for each experiment so that interference with cloud droplet sizes is avoided. Figure 2, row 4 shows the size distribution over time of growing droplets and ice crystals from Welas1 and Welas2. The size threshold is set to \( d_p = 30 \mu m \) in this experiment.

4. Experimental results

We performed 18 immersion-freezing experiments with the desert dusts described in section 2c in the temperature range between \(-10^\circ \text{C}\) and \(-28^\circ \text{C}\). The pressure \( p_0 \), temperature \( T_0 \), total particle number concentration \( N_{tot} \), and total particle surface area \( S_{tot} \) prior to the start of each experiment are listed in Table 2.

Figure 3 depicts the ice-active particle fractions \( f_i \) versus temperature. We calculate \( f_i \) from the ice number concentration \( N_i \) from Welas2 (cf. section 2b) divided by \( N_{tot} \) for a time interval when \( N_i \) is still increasing and the loss of ice crystals due to sedimentation is negligible. The time range considered for the analysis is set for each experiment individually. For experiment ACI04_22 the considered time range is 75–450 s (cf. Fig. 2, row 3). Note that the calculated \( N_i \) was averaged over a time period of minimum 60 s as described in section 2b, resulting in up to five values of \( f_i \) per experiment.

The vertical error bars in Fig. 3 represent a factor of 2 estimated uncertainty in \( f_i \). This includes mainly the uncertainty of the number concentration of large ice crystals and the uncertainty of the aerosol number concentration.

In general \( f_i \) increases exponentially with decreasing temperature. Ice nucleation was first observed at a temperature of about \(-13^\circ \text{C}\) in the case of SD, and at around \(-16^\circ \text{C}\) in the case of ID. No ice was detected in the AIDA experiments with AD in the temperature range between \(-10^\circ \text{C}\) and \(-19^\circ \text{C}\). The ATD and CID samples were not examined in the whole temperature range from \(-10^\circ \text{C}\) to \(-28^\circ \text{C}\). The ATD and CID data points each were derived from a single experiment. Also, \( f_i \) is \( 4 \times 10^{-3} \) for ATD at about \(-21^\circ \text{C}\) and \( 1 \times 10^{-4} \) for CID at about \(-23^\circ \text{C}\). The largest activated fraction of all immersion-freezing experiments shown in this article was about 1%. This was reached in the case of AD, ID, and SD at a temperature of about \(-28^\circ \text{C}\).

a. Surface size distributions

An SMPS and an APS (section 2b) were applied to determine the number size distribution before each experiment run with a freshly dispersed dust sample. To compare the results of the two instruments, the aerodynamic diameter measured by the APS and the equivalent mobility diameter measured by the SMPS were converted into a volume-equivalent sphere diameter \( d_p \). For this conversion, the bulk density and the shape factor must be known (Hinds 1999). Typical densities used in dust aerosol studies range from 2.5 g cm\(^{-3}\) for sand (Davies 1979; Linke et al. 2006) to 2.65 g cm\(^{-3}\) for quartz (Tegen and Fung 1994; Hinds 1999; Kandler et al. 2007). Hinds (1999) tabulates a dynamic shape factor of 1.36 for quartz and 1.57 for sand. We applied a bulk density of 2.6 g cm\(^{-3}\) for all dust samples and found the best match between SMPS and APS for dynamic shape factors between 1.1 and 1.4. The surface size distributions were calculated from the number size distributions, assuming that the dust particles are spheres with the respective volume-equivalent sphere diameter. Figure 4 shows surface size distributions for all freezing experiments listed in Table 2. Note that the surface size distribution varies among the same dust. A lognormal distribution (1–2 modes) was then fitted to the data with median diameters \( d_{s,median} \) and geometric standard deviations \( \sigma \) as listed in Table 3. The sum of the area under the curve corresponds to \( S_{tot} \) available in the experiment. The value of \( S_{tot} \) is used in section 4b to calculate the ice-active surface site density \( n_s \), which is the number of ice-active surface sites per unit area.

b. Parameterization of immersion freezing

To parameterize our experimental results on immersion freezing, we used the so-called ice-active surface site density approach as already introduced and discussed by C09. This concept, which was also discussed and used by Niedermeier et al. (2010), Lüönd et al. (2010), and...
Murray et al. (2011), assumes that localized sites with specific activation energies for ice nucleation are equally distributed over the dust particle surface area. It is further assumed that different sites exist with a wide range of freezing probabilities as a function of temperature and time. From a large number of AIDA cloud expansion studies we have evidence that the formation of ice is mainly related to the total temperature change and the particle surface area [see Eq. (5) in C09]. Because of this observation, we suggest the ice-active surface site density formulation as a parameterization for immersion freezing on desert dust particles for the use in atmospheric models.

According to Eq. (9) in C09, the ice crystal number concentration formed by active IN in size bin $j$ ($N_{ij}$) is expressed as

$$N_{ij} = N_{\text{tot},j} \{1 - \exp[-S_{\text{ac},j}(T)]\},$$

where $N_{\text{tot},j}$ is the total number of particles in size bin $j$ and $S_{\text{ac},j}$ is the individual particle surface area in that size.
The density of ice-active surface sites \( n_s(T) \) is considered constant throughout the size distribution, assuming that the aerosol composition and surface properties do not vary with size.

For a polydisperse aerosol sample, we sum over all \( n \) size bins:

\[
\sum_{j=1}^{n} N_{i,j} = \sum_{j=1}^{n} N_{\text{tot},j} \{ 1 - \exp[-S_{\text{ac},j} n_s(T)] \}. \tag{2}
\]

For \( S_{\text{ac},j} n_s(T) \ll 1 \), Eq. (2) can be approximated by

\[
\sum_{j=1}^{n} N_{i,j} \approx \sum_{j=1}^{n} N_{\text{tot},j} S_{\text{ac},j} n_s(T) = n_s(T) \sum_{j=1}^{n} N_{\text{tot},j} S_{\text{ac},j}. \tag{3}
\]

Rearranging Eq. (3) yields

\[
n_s(T) = \frac{\sum_{j=1}^{n} N_{i,j}}{\sum_{j=1}^{n} N_{\text{tot},j} S_{\text{ac},j}} = \frac{N_i}{S_{\text{tot}}}. \tag{4}
\]

where \( S_{\text{tot},j} \) is the total surface area per unit volume of particles in a size bin and \( S_{\text{tot}} \) is the total surface area of particles in all \( n \) size bins.

We measure \( N_i \) in the AIDA experiment (cf. section 2b), and \( S_{\text{tot}} \) (section 4a) is measured prior to each cloud expansion experiment and multiplied by the pressure dilution \( p/p_0 \) to reduce it through the experiment. Figure 5 shows \( S_{\text{tot}}, N_i \), the droplet number concentration \( N_d \), and the calculated value of \( n_s \) versus decreasing temperature as an example for experiment ACI04_19 with AD. Note that \( N_i \) is small compared with \( N_d \) (e.g., \( N_i \approx 10^9 \text{ m}^{-3} \) and \( N_d \approx 10^8 \text{ m}^{-3} \) at \(-28^\circ \text{C}, \) corresponding to a frozen fraction \( f_i \) of about 1%), and \( S_{\text{tot}} \) decreases from initially \( 1.47 \times 10^{-4} \text{ m}^2 \text{ m}^{-3} \) at \(-23.5^\circ \text{C} \) (start of ice formation) to \( 1.17 \times 10^{-4} \text{ m}^2 \text{ m}^{-3} \) at \(-28^\circ \text{C}. \)

This calculation was repeated for all 16 freezing experiments. The resulting \( n_s \) values versus decreasing temperature are shown in Fig. 6. Note that the calculated \( n_s \) was averaged over a time period of minimum 60 s as described in section 2b. The plot also includes the corrected

<table>
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<th>Table 3. List of lognormal fit parameters to the surface size distributions (one or two modes).</th>
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<tr>
<td>Expt No.</td>
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<tr>
<td>ACI04_13 AD</td>
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<tr>
<td>ACI04_16 AD</td>
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<td>ACI04_19 AD</td>
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<tr>
<td>ACI04_04 SD</td>
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<tr>
<td>ACI04_37 SD</td>
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<td>ICIS07_17 ID</td>
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<td>ACI04_22 ID</td>
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<td>ICIS07_03 ATD</td>
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</table>

Fig. 5. Measured droplet and ice number concentration, particle surface area, and calculated ice-active surface site density for experiment ACI04_19 with Asian dust vs temperature.
surface site densities obtained by C09. A reanalysis of some of the older AIDA experiments showed that C09 overestimated the surface site density by about a factor of 10, mainly because of a systematic error, resulting in underestimating the aerosol surface area by this factor.

The new data points from this study suggest that an exponential function fits the data best. We fitted our new data points, not including the C09 data. Also the ATD data point was left out, since ATD is a processed and milled sample (see Möhler et al. 2006). Crushed particles from this manufacturing process probably involve a higher density of surface defects and therefore a higher ice activity.

The exponential fit is given by the equation

\[ n_s(T) = \exp\left[-0.517(T - 273.15) + 8.934\right] \text{[m}^{-2}] \]  

This equation can be used in models as a parameterization of immersion freezing on desert dust particles in the temperature range \(-12^\circ\) to \(-36^\circ\)C. It will be used and compared with other concepts in sections 5b, 5d, and 5e.

The ice-active fraction in a size bin \(f_{i,j}\) can be calculated via [see Eq. (1)]

\[ f_{i,j} = \frac{N_{i,j}}{N_{\text{tot},j}} = 1 - \exp\left[-S_{\text{a},j}n_s(T)\right] \approx S_{\text{a},j}n_s(T). \]  

The approximation is valid for small aerosol particles and experiments at higher temperatures as demonstrated in Fig. 7. However, for particles larger than about 3 \(\mu\)m, for example, and temperatures lower than \(-30^\circ\)C, \(f_i\) approaches one and has to be calculated with the exact formulation for each size bin.

5. Saharan dust event of May 2008

In the following we show an application of the immersion freezing parameterization. This will then be compared with measurements of IN at the Taunus Observatory on Mount Kleiner Feldberg.

a. Description and simulation with COSMO-ART

In May 2008 a major dust outbreak over the Sahara occurred. This event transported large Saharan dust amounts across the Mediterranean area and western Europe. We used the regional-scale online coupled model system COSMO-ART (Vogel et al. 2009; Bangert et al. 2011a) to simulate the development and the spatial distribution of the dust plume. The performance of the model system has been recently evaluated by Knote et al. (2011) under a broad range of conditions, and by Stanelle et al. (2010) and Bangert et al. (2011b) for case studies of Saharan dust outbreaks. COSMO-ART includes a detailed dust emission scheme (Stanelle et al. 2010; Vogel et al. 2006) that gives size-dependent emission fluxes of particles depending on the friction velocity and a dataset of surface properties (Marticorena et al. 1997; Callot et al. 2000). COSMO-ART uses a modal approach to simulate the size distributions of particle number, surface area, and mass. Three lognormal modes that differ in their number median diameter (mode 1: 0.64 \(\mu\)m; mode 2: 3.45 \(\mu\)m; mode 3: 8.67 \(\mu\)m) and geometric standard deviation (mode 1: 1.7; mode 2: 1.6; mode 3: 1.5) are applied.

In this application of the model system, we simulated dust only and neglected all other aerosol types. The horizontal spatial resolution was 0.25 \(\times\) in both directions, which corresponds to about 28 km. (The model domain is shown in Fig. 11). Forty vertical layers up to a height

![Fig. 6. The black solid line is a T-dependent fit to all natural dusts from this study (\(r^2 = 0.91\)). The plot also includes the data from C09 reduced by a factor of 10 (see section 4b).](#)

![Fig. 7. The fraction of ice vs the diameter of monodisperse aerosol calculated using Eq. (6) for different temperatures.](#)
of 20-km altitude were used. The simulation started at 0000 UTC 22 May 2008 and ended at 1700 UTC 31 May 2008. The model run was driven by the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System (IFS) boundary conditions. Those boundary conditions are updated every 6 h. At the beginning of our simulation we assumed the atmosphere to be dust-free. During the simulation period the dust emissions lead to the development of a significant dust plume. A comparison with observations demonstrated that the model system simulates the meteorological situation and the dust plume reasonably well (Bangert et al. 2011b).

Figure 8 shows a longitudinal vertical cross section of the simulated concentration of dust particles at 50°13’N (along the Taunus Mountains, Germany) at 1200 UTC 28 May 2008. The dust plume is about 800 km wide and extends from the surface to 8–11 km in height. The number concentration reaches its maximum of $1.11 \times 10^5$ L$^{-1}$ at around 3 km. The corresponding vertical cross section of the total surface area of dust particles (not shown here) gives a maximum surface area of $4.18 \times 10^7$ µm$^2$ L$^{-1}$ at around 3-km height. The total dust particle surface area and the temperature from the COSMO-ART model simulation served as an input for the calculation of the IN number concentration using our new parameterization.

Figure 9 compares the simulated mass concentration of particles less than 10 µm in diameter (PM10) from COSMO-ART (PM10 COSMO) and the Dust Regional Atmospheric Model (DREAM) (PM10 DREAM; Nickovic et al. 2001; Klein et al. 2010) to the measured PM10 mass concentration derived from APS size distributions (PM10 KF) during the Saharan dust event at the Taunus Observatory on Mount Kleiner Feldberg (cf. Fig. 7 in Klein et al. 2010). PM10 KF is affected by the background concentration of small (e.g., sulfate, nitrate, soot) and organic particles, which leads to higher particle number concentrations compared with PM10 COSMO and PM10 DREAM, which include dust only. The background mass concentration at the Taunus Observatory is on the order of 15–25 µg m$^{-3}$ (cf. Klein et al. 2010). PM10 COSMO and PM10 DREAM show an increase during the time of the dust event, however, and predict a factor of 2–4 higher PM10 mass concentrations than the measurements.

The surface area calculated from the COSMO-ART modeled dust particle size distribution (S COSMO) is compared to the surface area derived from APS size spectra (S KF) during the time of the Saharan dust event in Fig. 10. The peak values of S COSMO and S KF are 270 and 400 µm$^2$ cm$^{-3}$, respectively. The numbers are in good agreement; however, they are reached at different times. The peak in S COSMO is reached on 29 May 2008 and in S KF on 30 May 2008.

b. Application of new parameterization

Based on the model results from COSMO-ART (total dust particle surface area and temperature) we applied the new parameterization [see Eqs. (4) and (5)] to calculate the spatial distribution of immersion freezing nuclei at 6000 m above surface (Fig. 11), independent of the simulated relative humidity. At 6000-m height, temperatures around −18°C were reached. The pattern shows high IN concentrations of more than 500 L$^{-1}$ above northern Africa and northeast of Spain and spreading across western Europe.
Europe. The black line in Fig. 11 indicates the location of the vertical cross section at 50°13’N presented in Figs. 8 and 12.

Figure 12 shows the vertical distribution of potential immersion freezing nuclei at this latitude. For this calculation we used a constant temperature of −18°C for comparison with the Frankfurt Ice Deposition Freezing Experiment (FRIDGE; Bundke et al. 2008) IN measurements that were performed at this temperature (section 5c). The maximum ice number concentration of 27 L−1 is reached at 3-km height.

c. Measurements of IN at the Kleiner Feldberg field site

The number concentration of deposition/condensation freezing IN has been measured at Taunus Observatory on Mount Kleiner Feldberg since April 2008 on a regular daily basis. For these measurements, the atmospheric aerosol is collected for time periods of 5 min by electrostatic precipitation on silicon wafers in the 10-nm–20-μm size range. The samples are then analyzed in FRIDGE at −18°C and RH_{ice} = 119% (RH_{water} ≈ 100%) for their IN number. The grown ice crystals are viewed by a charge-coupled device (CCD) camera and counted automatically. High aerosol concentrations and a strong increase in the IN number concentrations at −18°C were measured at the Taunus Observatory during the Saharan dust event in May 2008 (Klein et al. 2010). In addition, an APS from TSI, Inc., was used to measure aerosol particle number distributions in the size range 0.5–20 μm during the time of the dust event. The aerosol surface area calculated from the APS size distributions was used to calculate the IN number concentration with the new parameterization. We compare the results to the IN number calculated from the simulation and to FRIDGE measurements in section 5d.

d. Comparison of modeled and measured results

Here we compare the number concentrations of calculated IN to those measured with FRIDGE (section 5c) for a temperature of −18°C (Fig. 13). A background concentration of 26.27 L−1 (average IN number concentration in FRIDGE before and after the dust event)
was subtracted from the measurements during the dust event.

Because RHwater is approximately 100% in FRIDGE, it can be assumed that water uptake is involved in the freezing process. Most of the ice formation in the AIDA cloud chamber experiments of this work occurred through the immersion freezing mode, with some minor contribution of condensation freezing close to the onset of cloud condensation. In a comparison study by DeMott et al. (2011), FRIDGE and AIDA showed similar freezing onsets in the case of ATD, but it should be mentioned here that we cannot yet completely rule out a systematic difference between both methods, in particular if condensation and immersion freezing rates and active fractions markedly differ from each other at −18°C.

For the comparison with FRIDGE measurements, the simulated dust concentration at the closest point to Mount Kleiner Feldberg (model layer at 800 m ASL) was taken as a basis. The gray shaded area in Fig. 13 indicates the uncertainty in the calculated IN number concentration due to the uncertainty in the simulated particle surface area (section 5d).

Figure 14 shows our new parameterization in comparison to three other concepts, including the parameterizations by DeMott et al. (2010), Hoose et al. (2010), and Phillips et al. (2008) for the temperatures −15°C, −18°C, and −25°C. The total particle surface area or number concentrations calculated from the COSMO-ART model simulation are used as input parameters for the different parameterizations.

Whereas our parameterization is based on laboratory studies in the AIDA cloud chamber (IN number concentration as a function of dust particle surface area and temperature), the parameterizations by DeMott et al. (2010) and Phillips et al. (2008) are based on field measurements of IN number concentrations with a continuous flow diffusion chamber. In the parameterization by DeMott et al. (2010), the IN number concentration is a function of number concentration (dp > 0.5 μm) and temperature. The parameterization by Phillips et al. (2008) uses the particle surface area and the ice supersaturation as input parameters. Phillips et al. (2008) extrapolated their fit to temperatures warmer than −25°C. The temperature-dependent parameterization by Hoose et al. (2010) is based on classical nucleation theory and laboratory experiments with montmorillonite/illite. The parameterizations by Hoose et al. (2010) and Phillips et al. (2008) are both evaluated at water saturation. Note that all parameterizations, except the one by DeMott et al. (2010), are proportional to the particle surface area and differ only by a constant factor at a constant temperature.

Our parameterization is most sensitive to the temperature variation. The peak IN number concentrations for −15°C, −18°C, and −25°C are about 5, 22, and 742 L−1, respectively. The IN number concentrations calculated with the parameterization by DeMott et al. (2010) are roughly 3 L−1 (−15°C), 7 L−1 (−18°C), and 36 L−1 (−25°C). The parameterization by Phillips et al. (2008) gives IN number concentrations of about 227 L−1 (−15°C), 357 L−1 (−18°C), and 1068 L−1 (−25°C). The parameterization by Hoose et al. (2010) gives values of about
The parameterization by Hoose et al. (2010) leads to higher values of IN because of the time dependency and the integration over 90 s, which corresponds to the measurement time of FRIDGE. Furthermore, this parameterization is based on experiments with montmorillonite/illite, pure clay minerals that are known to be very efficient IN. The extrapolation of Phillips et al. (2008) to temperatures warmer than $-18^\circ$C is uncertain because of the lack of constraining data. They used the form of the parameterization by Meyers et al. (1992) as a basis for their extrapolation. The parameterization shows a relatively weak dependence on temperature between $-15^\circ$ and $-25^\circ$C. The lower IN number concentrations predicted by the DeMott et al. (2010) parameterization are likely due to its derivation from CFDC measurements sampling background concentrations in primarily non-dust-dominated environments.

6. Summary and atmospheric implications

Immersion freezing on different desert dust particles was investigated in the AIDA cloud expansion chamber at temperatures between $-12^\circ$ and $-28^\circ$C. We derived a parameterization as a function of the dust particle surface area and temperature that fits the measured abundance of immersion freezing nuclei in desert dust aerosols. The parameterization can be applied in atmospheric models that include information about the dust surface area.

Based on the surface area calculated from the COSMO-ART-modeled dust particle size distribution and the COSMO-ART-modeled temperature distribution, the new parameterization was applied to calculate spatial distributions of IN number concentration during a large Saharan dust outbreak in May 2008. The results were compared to size distributions and to IN number concentrations measured at the Taunus Observatory on Mount Kleiner Feldberg in Germany for a temperature of $-18^\circ$C and a relative humidity with respect to ice of 119% ($\text{RH}_{\text{water}} \approx 100\%$). The dust surface area from the model simulation agreed well with the aerosol surface area calculated from the measured aerosol size distributions, but the calculated IN number concentrations were about 13 times lower than the measured ones.

We also compared our parameterization to three other approaches, namely the parameterizations by DeMott et al. (2010), Hoose et al. (2010), and Phillips et al. (2008) at three different temperatures based on the aerosol number concentration and surface area from the COSMO-ART model simulation. Our parameterization is most sensitive to temperature variations. It gives results similar to the parameterization by DeMott et al. (2010) for a temperature of $-15^\circ$C. For $-18^\circ$C the parameterizations by
Phillips et al. (2008) and Hoose et al. (2010) give IN number concentrations that are 16−51 times higher compared with our parameterization but are in better agreement with the FRIDGE measurements. For −25°C our parameterization gives about a factor of 4 less IN than the parameterizations by Hoose et al. (2010), but 21 times higher IN number concentrations than the parameterization by DeMott et al. (2010).

There are several reasons that could cause the discrepancies between the calculated and measured IN number concentrations and systematic differences among the different parameterizations. Uncertainties in the modeled and measured parameters both contribute to the discrepancy between calculated and measured IN number concentrations. For example, biological particles attached to the transported dust could lead to a higher IN number concentration (Conen et al. 2011), and local dust emissions could lead to an enhanced aerosol surface area compared to the calculations using the modeled Saharan dust concentration only. Concerning the accuracy of IN number concentration measurements, DeMott et al. (2011) published a comparison between different methods and instruments for IN measurements. This comparison shows that FRIDGE measures ice nucleation onsets similar to those of other instruments, but comprehensive comparisons of measured IN number concentrations have not been obtained. Future comparisons of IN measuring devices are therefore merited.

Also, care has to be taken when comparing parameterizations derived from experiments with different dust samples. There are indications that milled and processed samples such as ATD and illite have a higher ice nucleation efficiency than natural dust samples (Möhler et al. 2006), which could explain at least part of the discrepancy between different parameterizations based on laboratory measurements. To further resolve the discrepancies between parameterizations and field measurements, more quantitative intercomparison studies between in situ IN measurements and model studies are needed.

It remains to be examined whether the surface soil samples are representative for long-range transported dust, because during the transport the aerosols are chemically processed and thereby modified concerning the ice nucleation efficiency. Laboratory studies (Möhler et al. 2008; Eastwood et al. 2009; Cziczo et al. 2009; Niedermeier et al. 2010; Sullivan et al. 2010) and field experiments (Ansmann et al. 2005) have shown that chemical processing can in some cases lower the IN ability. It is a subject of ongoing work to quantify the effect of various surface coatings on the ice-active surface site density on mineral dust particles.

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