A New Ice Nucleation Active Site Parameterization for Desert Dust and Soot

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ABSTRACT

Based on results of 11 yr of heterogeneous ice nucleation experiments at the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) chamber in Karlsruhe, Germany, a new empirical parameterization framework for heterogeneous ice nucleation was developed. The framework currently includes desert dust and soot aerosol and quantifies the ice nucleation efficiency in terms of the ice nucleation active surface site (INAS) approach.

The immersion freezing INAS densities \( n_S \) of all desert dust experiments follow an exponential fit as a function of temperature, well in agreement with an earlier analysis of AIDA experiments. The deposition nucleation \( n_S \) isolines for desert dust follow u-shaped curves in the ice saturation ratio–temperature \((S_i–T)\) diagram at temperatures below about 240 K. The negative slope of these isolines toward lower temperatures may be explained by classical nucleation theory (CNT), whereas the behavior toward higher temperatures may be caused by a pore condensation and freezing mechanism. The deposition nucleation measured for soot at temperatures below about 240 K also follows u-shaped isolines with a shift toward higher \( S_i \) for soot with higher organic carbon content. For immersion freezing of soot aerosol, only upper limits for \( n_S \) were determined and used to rescale an existing parameterization line.

The new parameterization framework is compared to a CNT-based parameterization and an empirical framework as used in models. The comparison shows large differences in shape and magnitude of the \( n_S \) isolines especially for deposition nucleation. For the application in models, implementation of this new framework is simple compared to that of other expressions.

1. Introduction

Ice particles in clouds influence both the radiation budget of Earth and the lifetime of clouds and often initiate the formation of precipitation. In the troposphere, they are formed by either homogeneous freezing or heterogeneous nucleation processes. Homogeneous freezing happens at temperatures below about 238 K and high ice saturation ratios that are usually just reached in the upper troposphere. Heterogeneous ice nucleation is controlled by the abundance and type of solid aerosol particles. Well below water saturation, deposition nucleation occurs if water vapor directly deposits on the aerosols surface. Near water saturation, water can condense in pores and cavities, and subsequently freezes, if cold enough, by the pore condensation and freezing (PCF) mechanism (Marcolli 2014; Wagner et al. 2016). Above water saturation, supercooled droplets are formed and can freeze subsequently, either induced by a solid aerosol particle inside the droplet (immersion freezing), or an aerosol particle colliding with the droplet (contact nucleation) (Vali et al. 2015).

Aerosol particles in the atmosphere originate from different sources and include a variety of compounds. The most abundant particle types are composed of sulfate, mineral dust, sea salt, soot, and organic material (Stier et al. 2005). However, only a minor part of the atmospheric aerosol particles contributes to the ice formation in clouds. The ice active particle fraction strongly depends on the aerosol type and the temperature (Hoos and Möhler 2012; Murray et al. 2012). Mineral dust was found to be a major compound in residual particles of cirrus ice particles formed by heterogeneous ice nucleation (Cziczo et al. 2013; Kamphus et al. 2010). Signatures from biomass burning and sea salt aerosols as well as mixtures of sulfate, organics, and nitrate were also found in the cirrus ice residuals, depending on the geographic region and season.
The Fifth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC AR5; Boucher et al. 2013) and Komurcu et al. (2014) showed that the simulated ice clouds in global climate models are very sensitive to the implemented parameterization of the aerosol's ice-nucleating ability. The early descriptions for heterogeneous ice nucleation by Fletcher (1962) or Meyers et al. (1992) are neither aerosol specific nor applicable over the whole tropospherically relevant temperature and humidity range. The first ice-nucleating particle (INP) parameterization frameworks meeting these conditions were derived from field and laboratory measurements (Phillips et al. 2008, 2013) or were based on classical nucleation theory (CNT) fitted to laboratory data (e.g., Hoose et al. 2010; Savre and Ekman 2015).

The paper here comprises a parameterization framework for the heterogeneous ice nucleation of desert dust and soot aerosol only depending on the aerosol surface area distribution, the temperature, and the ice saturation ratio. The framework is based on the database of laboratory measurements with the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) cloud chamber. It follows the ice nucleation active surface site (INAS) density concept, which is based on the early work of Vali (1971) and assumes that the freezing of supercooled droplets is to a first approximation time independent and that the ice germ formation is induced by specific sites on the aerosol surface. The INAS density $n_S$ for a polydisperse aerosol sample is given by

$$\sum_{j=1}^{k} n_{i,j}(T, S_i) = \sum_{j=1}^{k} n_{ac,j} \{1 - \exp[-S_{ac,j} n_S(T, S_i)]\}, \quad \text{(1)}$$

where $n_{i,j}$ is the ice number concentration formed on the aerosol, $n_{ac,j}$ the total aerosol number concentration, and $S_{ac,j}$ the individual aerosol surface area in size bin $j$. A detailed derivation can be found in, for example, Vali (1971), Connolly et al. (2009), or Niemand et al. (2012).

This study is based on data measured with the AIDA cloud chamber, which will briefly be described in section 2a. Sections 2b and 2c will document the data processing and evaluation and will give an overview of the aerosol samples used for the AIDA experiments. The INAS density results for immersion freezing and deposition nucleation, and our new parameterizations will be presented in section 3. In section 4, the potential use of the parameterization in models will be discussed.

### 2. Data and methods

All experiments were performed in the AIDA cloud chamber at the Karlsruhe Institute of Technology. AIDA can be operated as a cloud simulation chamber to investigate the formation of clouds and, in particular, the primary ice formation inside clouds under realistic atmospheric conditions.

#### a. Experimental setup

The core of the AIDA cloud chamber is an 84-m$^3$-sized aluminum vessel in which the ascent of a moist air parcel in the atmosphere is simulated by evacuating the vessel. After some time of expansion cooling, the air inside AIDA exceeds saturation with respect to ice and/or water so that the present aerosol particles can act either as INPs and/or as cloud condensation nuclei (CCN) to form pure ice clouds, pure supercooled liquid clouds, or mixed-phase clouds. A more technical description of the AIDA cloud chamber and its instrumentation can be found in, for example, Möhler et al. (2005a, 2006), Benz et al. (2005), or Wagner et al. (2012).

Figure 1 shows the ice number concentrations and the total particle concentration for a typical (Fig. 1a) immersion freezing and (Fig. 1b) deposition nucleation experiment. The ice number concentration is measured by two optical particle counters Welas 1 and 2 (Palas GmbH) (Benz et al. 2005). The detection range of Welas is 0.7–46 μm and of Welas is 2.5.0–240 μm (Wagner and Möhler 2013). With start of the expansion, the temperature decreases and the ice saturation ratio increases. For specific thermodynamic conditions, ice starts to form indicated by the first red vertical line. The second red vertical line indicates the stop of new ice formation. The differences between the Welas 1 and 2 ice number concentrations are caused by the different detection ranges.

#### b. Data processing

In this study, the ice nucleation efficiency of the aerosol samples is quantified by the INAS density, calculated by dividing the ice number concentration $n_i$ by the aerosol surface area concentration $n_{ac}$, assuming that $n_{ac} n_S \ll 1$ [see Eq. (1)] and that $n_S$ is constant throughout the aerosol size distribution

$$n_S(T, S_i) = \frac{n_i(T, S_i)}{n_{ac}(S_{ac})} = \frac{n_i(T, S_i)}{S_{ac}}. \quad \text{(2)}$$

Because aerosol, droplets, and ice crystals are measured in different optical diameter ranges, a diameter threshold is set to calculate the ice number concentrations from Welas 1 and Welas 2. A time interval is set (vertical red dashed lines in Fig. 1) in order to limit the analysis of the ice nucleation to the period of valid and most accurate data. The start of this interval is the time when the ice number concentration exceeds its background value of about 0.1 cm$^{-3}$. The end of the interval is...
reached when the ice formation rate drops to zero, which is indicated by the decreasing slope of ice number concentration.

This ice nucleation time interval is further split into smaller time bins for the analysis. To obtain representative statistics, each bin \( k \) is required to have a minimum length of 10 s and to include at least five ice particle counts. For each bin, the ice number concentration \( n_{i,k} \), the mean temperature \( T_k \), and the mean ice saturation ratio \( S_{i,k} = \frac{RH_{i,k}}{100\%} \) are determined. For immersion freezing experiments, \( n_i \) is calculated from the Welas 2 instrument, because of its larger detection volume and therefore higher sensitivity to measure low ice particle concentrations. The Welas 1 data are used for deposition nucleation experiments at low temperatures, because in this temperature regime ice crystals grow more slowly, therefore stay smaller in size, and can be detected earlier and more sensitively in the smaller size range of Welas 1.

The aerosol size distribution is determined from combined measurements of a Scanning Mobility Particle Sizer (SMPS, TSI) and an Aerodynamic Particle Sizer (APS, TSI) (Niemand et al. 2012). The total aerosol number and surface area concentrations are obtained from a lognormal fit to the size distribution. The pumping during the experiment leads to a reduction of the aerosol concentration. Therefore, the total number and surface area concentrations as measured prior to the pumping and are corrected time-bin-wise by the pressure fraction \( \frac{p_k}{p_0} \), where \( p_k \) is the mean pressure in bin \( k \) and \( p_0 \) the pressure at start of the expansion. For immersion freezing, only the aerosol activated to droplets will contribute to ice formation. If the CCN activated fraction is less than 50% the total aerosol surface area concentration is reduced to this fraction (see appendix C, section a), otherwise the correction is negligible and not considered. The aerosol surface area concentration \( s_{ae,k} \) in each time bin \( k \) is the fraction of the total aerosol surface area concentration \( s_{ae,0} \), which, until this time, was activated to ice crystals. This fraction is obtained by integrating the lognormal distribution from a threshold particle diameter \( d_{act,k-1} \) to infinity and corrected for the dilution effect

\[
s_{ae,k} = s_{ae,0} \frac{p_k}{p_0} \Phi \left( \frac{\log d_{act,k-1} - \log d_{m,s}}{\log \sigma_{g,s}} \right).
\]

By doing so, we imply that the largest aerosol particles activate first (see appendix C, section b, for a detailed derivation). This assumption only is a first-order correction taking into account that the nucleation probability is proportionate to the squared particle diameter. In Eq. (3), \( \Phi(x) \) is the standard normal distribution function with the cutoff value \( x \), \( d_{m,s} \) is the median diameter, and \( \sigma_{g,s} \) the geometric standard deviation of the aerosol surface area size distribution function, and \( d_{act,k-1} \) is the aerosol diameter above which the aerosols were activated in the previous time bin calculated from the ice number concentration.

Following the described algorithm, the INAS density for each bin \( k \) is then calculated for each bin \( k \) by

\[
n_{S}(T, S_{i,k}) = \frac{n_{i,k}(T, S_{i,k})}{s_{ae,k}}.
\]

Note that this approach is different from the cumulative INAS density calculation by, for example, Niemand et al.
Table 1. Labels and origin of the desert dust samples.

<table>
<thead>
<tr>
<th>Sample type</th>
<th>AIDA label</th>
<th>Sample description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Asian desert dust</td>
<td>AD1, AD2</td>
<td>Collected from the eastern parts of the Takla Makan desert in China, east of the Dalim basin between Kuerle and Ruoquiang (Möhler et al. 2006)</td>
</tr>
<tr>
<td>Canary Island dust</td>
<td>CID1</td>
<td>Collected from the ground near Mala, Lanzarote, Canary Islands (Kanji et al. 2011)</td>
</tr>
<tr>
<td>Israel desert dust</td>
<td>ID1</td>
<td>Collected as sediment particles after a Saharan dust storm event in Tel Aviv, Israel (Kanji et al. 2011)</td>
</tr>
<tr>
<td>Saharan desert dust</td>
<td>SD2</td>
<td>Collected from a hole of 1.5-m depth about 70 km northeast of Cairo, Egypt (Megahed 2007, therein labeled as SD)</td>
</tr>
<tr>
<td></td>
<td>SD1, SD4</td>
<td>Collected 50 km north of Cairo, Egypt as sediment particle after sand storm event (Megahed 2007, therein labeled as CD1 and CD2) (Kanji et al. 2011)</td>
</tr>
<tr>
<td></td>
<td>SD6</td>
<td>Collected in Morocco</td>
</tr>
<tr>
<td></td>
<td>SD19</td>
<td>Collected as soil sample in southern Tunisia near Tozeur 33°58′53.93″N, 8°0′6.72″E (E. Toprak 2013, personal communication)</td>
</tr>
</tbody>
</table>

(2012) or Steinke et al. (2011). The error propagation with uncertainties of $\Delta n_{i,k} = 0.2$ (Wagner and Möhler 2013) and $\Delta n_{\text{se},k} = 0.34$ yields a relative error for the INAS density of about 40%.

c. Origin, preparation, and characterization of the aerosol samples

1) Desert Dust

The desert dust samples in this study originate from seven different locations around the world and were collected from the ground (see Table 1). The sample SD1 was collected as sedimented particles 50 km north of Cairo City during a sand storm event on 18 February 2003 (Megahed 2007, labeled as CD1 therein). With a back-trajectory analysis, Megahed (2007) showed that the origin of sample SD1 is the Sahara desert. SD4 was collected at the same location as sample SD1 (Kanji et al. 2011; Niemand et al. 2012). In contrast, SD2 was sampled 1.5 m below ground. All samples were sieved to remove particles larger than 75 μm. Before injecting the aerosol into the AIDA cloud chamber, most of the samples passed a cyclone impactor with a cutoff value between 1 and 5 μm to further remove larger particles.

Elemental analysis of the sieved samples SD1, SD2, SD4, AD1, and AD2 showed strong signals of silicon dioxide indicating that the samples contain quartz (Megahed 2007; Möhler et al. 2006). For SD2, Linke et al. (2006) (therein labeled as Cairo2) have not been able to detect iron oxides by using x-ray diffractionometry (XRD). By ion-chromatographic analysis of aqueous extracts of SD1, SD2, SD4, AD1, and AD2, Megahed (2007) found that the samples contain less than 0.6 percent by weight (wt%) of ionic species indicating that the samples may contain a very low amount of anthropogenic pollutants. The samples ID1 and CID1 were collected after Saharan dust storm events (Kanji et al. 2011; Koehler et al. 2010). Koehler et al. (2010) suggested that the CID1 sample may contain beach sand and small amounts of ancient coral. All samples were considered to provide a representative spectrum of natural desert dust.

2) Soot

The soot samples used in this study were generated with four devices. A Combustion Aerosol Standard (CAST) burner (Jing Technologies) was used to generate soot in a coflow diffusion of the fuel gas propane and synthetic air (Möhler et al. 2005a, their Fig. 1). The controlled variation of the propane-to-air ratio (and therewith the C/O ratio; Schnaiter et al. 2006) determines the organic-to-elemental carbon (OC–EC) ratio of the soot. A modified version of the CAST burner, the so-called mini CAST burner (mCAST), operates in a similar way, but generates soot with a somewhat higher OC-to-EC ratio for the same fuel C/O ratio (Crawford et al. 2011, their Fig. 3). A detailed description of the operating principle of the CAST and mCAST burner can be found in, for example, Möhler et al. (2005a), Crawford et al. (2011), and Schnaiter et al. (2006).

The third device is a graphite spark generator (GFG 1000, Palas GmbH). The soot from this device is formed from evaporated and recondensed graphite. The generated particles are transported within a carrier gas (argon or nitrogen), where they coagulate to larger agglomerates.

The diesel soot sample was generated with a diesel engine test bench (Laborde et al. 2012) with and without a particle filter. To reduce the concentration of water, volatile organics, and nitrogen oxides, the motor exhaust passed a series of denuders before it was added to the AIDA cloud chamber (Saathoff et al. 2003).

In the following, the soot aerosol is characterized for its organic carbon (OC) content using the thermographic carbon analysis according to the German VDI-guideline 2465, part 2 (VDI 1999). The OC mass content refers to the fraction of carbon which thermally desorbs at 650°C in a pure helium atmosphere (Schnaiter et al. 2006).
TABLE 2. Conditions for immersion freezing experiments with desert dust: \( nae,0 \) is the initial aerosol number concentration measured by the CPC; \( sae,0 \) is the initial aerosol surface area concentration; and \( T_{start} \), \( S_{start} \), and \( nS,\text{start} \) are the temperature, ice saturation ratio, and INAS density at start of ice nucleation, respectively. When two values are listed in the \( sae,0 \) column, the first value is the smaller mode and the second value is the larger mode of the aerosol surface area lognormal distribution. Previously published experiments are labeled as N12 (Niemand et al. 2012) and C09 (Connolly et al. 2009).

<table>
<thead>
<tr>
<th>Campaign</th>
<th>Expt No.</th>
<th>Date</th>
<th>Aerosol</th>
<th>( nae,0 ) (cm(^{-3}))</th>
<th>( sae,0 ) (( \mu \text{m}^2 \text{cm}^{-3} ))</th>
<th>( T_{start} ) (K)</th>
<th>( S_{start} )</th>
<th>( nS,\text{start} ) (m(^{-2}))</th>
<th>Published</th>
</tr>
</thead>
<tbody>
<tr>
<td>ACI04</td>
<td>7</td>
<td>30 Sep 2010</td>
<td>SD4</td>
<td>173.88</td>
<td>212.46</td>
<td>253.9</td>
<td>1.19</td>
<td>1.778 \times 10^8</td>
<td>N12</td>
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<tr>
<td>ACI04</td>
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<td>1.056 \times 10^8</td>
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</tr>
<tr>
<td>ACI04</td>
<td>16</td>
<td>1 Oct 2010</td>
<td>AD2</td>
<td>238.25</td>
<td>195.09</td>
<td>253.3</td>
<td>1.19</td>
<td>1.408 \times 10^8</td>
<td>N12</td>
</tr>
<tr>
<td>ACI04</td>
<td>19</td>
<td>4 Oct 2010</td>
<td>AD2</td>
<td>218.24</td>
<td>1.05, 186.14</td>
<td>249.3</td>
<td>1.27</td>
<td>1.719 \times 10^8</td>
<td>N12</td>
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<tr>
<td>ACI04</td>
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<td>ID1</td>
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<td>163.71</td>
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<td>385.56, 90.54</td>
<td>257.5</td>
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<td>ID1</td>
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<tr>
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<td>34</td>
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<tr>
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<td>805.49</td>
<td>255.4</td>
<td>1.17</td>
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<tr>
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<td>ID1</td>
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<td>252.3</td>
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<tr>
<td>ICIS07</td>
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<td>25 Sep 2007</td>
<td>SD4</td>
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<td>92.8</td>
<td>252.7</td>
<td>1.15</td>
<td>1.403 \times 10^8</td>
<td>N12</td>
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<tr>
<td>ICIS07</td>
<td>23</td>
<td>26 Sep 2007</td>
<td>CID1</td>
<td>381.81</td>
<td>58.68</td>
<td>250.3</td>
<td>1.18</td>
<td>1.228 \times 10^8</td>
<td>N12</td>
</tr>
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<td>IN04</td>
<td>6</td>
<td>17 Sep 2003</td>
<td>SD2</td>
<td>171.67</td>
<td>67.88</td>
<td>246.0</td>
<td>1.13</td>
<td>8.747 \times 10^7</td>
<td>C09</td>
</tr>
<tr>
<td>IN04</td>
<td>18</td>
<td>24 Sep 2003</td>
<td>AD1</td>
<td>181.33</td>
<td>72.49*</td>
<td>242.9</td>
<td>1.32</td>
<td>5.955 \times 10^7</td>
<td>C09</td>
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<tr>
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<td>29 Sep 2003</td>
<td>SD2</td>
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<td>127.93</td>
<td>249.0</td>
<td>1.11</td>
<td>3.736 \times 10^7</td>
<td>C09</td>
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<tr>
<td>IN19</td>
<td>23</td>
<td>16 Jul 2012</td>
<td>SD19</td>
<td>180.77</td>
<td>99.27</td>
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<td>2.530 \times 10^8</td>
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<tr>
<td>FIN02</td>
<td>3</td>
<td>14 Mar 2015</td>
<td>SD6</td>
<td>735.45</td>
<td>88.47, 25.71</td>
<td>249.1</td>
<td>1.23</td>
<td>1.880 \times 10^8</td>
<td>—</td>
</tr>
</tbody>
</table>

*The value of \( sae,0 \) was estimated from \( nae,0 \) and for typical \( d_{mean} \) and \( \sigma_{g,n} \).

3. Results

As mentioned in section 2, the ice nucleation ability is quantified by the INAS density. For the immersion freezing regime, previous studies (Connolly et al. 2009; Hoose and Möhler 2012; Niemand et al. 2012) introduced the INAS density as a function of the temperature. More recent studies (Hiranuma et al. 2014b; Steinke et al. 2015) extended this surface site approach to deposition nucleation, where the ice nucleation activity depends on both the temperature and the ice saturation ratio.

### a. Immersion freezing

#### 1) DESERT DUST

Table 2 lists all AIDA immersion freezing experiments on desert dust performed in the past 11 yr with their corresponding initial aerosol number and surface area concentration, the temperature, the ice saturation ratio, and the INAS density for the first analyzed time bin as described in section 2b. As we improved the evaluation algorithm in this study (see section 2b), also earlier published experiments from Niemand et al. (2012) and Connolly et al. (2009) were reanalyzed. Those experiments are marked in Table 2. For the experiments where no ice was detected (Table 3), the temperature at start of droplet formation and at stop of evacuation are listed instead.

Figure 2a shows the temperature-dependent INAS densities for desert dust aerosol from all experiments listed in Table 2. Note that, because of the time-bin-wise evaluation, one or more data points per experiment can be obtained. A typical error bar for the temperature and the INAS density is shown for only one data point.
Freezing of supercooled droplets was detected between 243 and 259 K. The calculated INAS densities range from about $4.0 \times 10^{10}$ to $1.0 \times 10^{7}$ m$^{-2}$, increasing for decreasing temperature. Following Niemand et al. (2012), the INAS density values derived from the measurements for immersion freezing on desert dust can be approximated by an exponential function (see Fig. 2a, dashed line):

$$n_S(T, \text{ desert dust}) = \exp(150.577 - 0.517T) \text{ m}^{-2},$$

$$T \in [243, 259] \text{ K}.$$

This function yields $n_S$ values ranging from $7.4 \times 10^{10}$ to $1.9 \times 10^{7}$ m$^{-2}$ in the temperature range from 243 to 259 K.

Comparing the different desert dust samples concerning their ice nucleation ability we did not observe large discrepancies, as also reported by Niemand et al. (2012). Note that this also holds for the samples that have been transported through the atmosphere, indicating that the INAS density fit line may be representative for immersion freezing of both freshly emitted and transported dust.

Table 3. Conditions for immersion freezing experiments with desert dust where no freezing was observed: $n_{ae,0}$ is the initial aerosol number concentration measured by the CPC, $s_{ae,0}$ is the initial aerosol surface area concentration, $T_{droplet}$ is the temperature at the start of droplet formation, and $T_{stop}$ is the temperature at which the expansion stopped. Previously published experiments are labeled with N12 (Niemand et al. 2012) and C09 (Connolly et al. 2009).

<table>
<thead>
<tr>
<th>Campaign</th>
<th>Expt No.</th>
<th>Date</th>
<th>Aerosol</th>
<th>$n_{ae,0}$ (cm$^{-3}$)</th>
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induced ice formation in previous time bins. In the current analysis, this procedure reduces the aerosol surface area and, therefore, increases the resulting INAS density compared to uncorrected data.

2) SOOT

So far, only a few experiments with soot were done in the immersion freezing regime. Heterogeneous ice formation was only observed in two experiments with CAST\_minOC soot (ACI03\_5 and IN09\_6) and an experiment with GSG soot (IN09\_3) (Table 4), but overall only five or fewer ice particles were counted. During five experiments with GSG soot (see Table 4), supercooled droplets formed at about 240 K, but only homogeneous freezing was observed upon further cooling to about 238 K. For the experiment HALO06\_2, supercooled droplets existed in the temperature range from 251 to 248 K, but no freezing was observed. Based on the Welas detection limit, only an upper limit for the INAS density value was determined for these experiments in contrast to the analysis of the previously shown desert dust experiments. These upper limits were calculated by taking a total number of five ice crystals as the Welas ice number detection limit in the whole time period with the supercooled droplet cloud present in the AIDA chamber.

Accordingly, Fig. 2b depicts one INAS density upper-limit value for each experiment together with literature data. Compared to the data points calculated by DeMott (1990), the \( n_s \) values from the AIDA measurements are up to three orders of magnitude lower. However, there is a very good agreement with the very recent study of Schill et al. (2016) using diesel exhaust particles. The AIDA measurements suggest that soot with a low OC content is a poor immersion freezing INP. The deviation between the laboratory results may indicate a highly variable ice nucleation activity of different soot types.

To estimate a parameterization line for the INAS density of soot, the parameterization given by Murray et al. (2012) was scaled to the AIDA data points in Fig. 2b, as done by Schill et al. (2016):

\[
\begin{align*}
    n_s(T, \text{soot}) &= 7.463 \exp\left(-0.0101(T - 273.15)^2\right) \\
    &\quad - 0.8525(T - 273.15) + 0.7667 \text{ m}^{-2}, \\
    T &\in [239, 255] \text{ K}.
\end{align*}
\]

This function yields \( n_s \) values ranging from \( 5.4 \times 10^8 \) to \( 3.0 \times 10^9 \text{ m}^{-2} \) in the temperature range from 239 to 255 K. Note that this function represents the upper-limit INAS density values derived from AIDA immersion freezing experiments with soot. Therefore, the parameterization might have to be adapted to even lower INAS density values when more experiments with soot in the immersion freezing mode are available for analysis.

3) COMPARISON TO OTHER IMMERSION FREEZING PARAMETERIZATIONS

Figure 3 shows a summary of the parameterization for desert dust and soot together with INAS density parameterizations from the literature. The lines for the dust components illite NX (Hiranuma et al. 2015), K-feldspar (Atkinson et al. 2013), and kaolinite (Murray et al. 2011) were scaled to their mean mass fraction in natural desert dusts following Atkinson et al. (2013, supplemental Table 1 therein). The line for milled hematite (Hiranuma et al. 2014a) was not scaled, because the mass fraction within natural desert dusts is unknown. Furthermore, note that illite NX, kaolinite, and K-feldspar are not pure samples but include some minor fraction of other minerals. Nevertheless, the scaled parameterization for illite NX does agree very well with the parameterization for desert dust. Therefore, illite seems to be a good proxy for ice nucleation of desert dust. Kaolinite, feldspar, and hematite are only minor components of natural desert dusts and show a much steeper decrease in INAS density with increasing temperature. When accounting for the

### Table 4. Conditions for immersion freezing experiments with soot: \( n_{s,0} \) is the initial aerosol number concentration measured by the CPC; \( s_{w,0} \) is the initial aerosol surface area concentration; \( n_{S,max} \) is the upper-limiting INAS density; \( T_{max} \) and \( S_{max} \) are the corresponding temperature and ice saturation ratio, respectively; and \( T_{droplet} \) is the temperature at start of droplet formation. If labeled with an asterisk, no upper-limiting \( n_s \) was determined.

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<th>( s_{w,0} ) (( \mu \text{m}^2 \text{ cm}^{-3} ))</th>
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different determination of the aerosol surface area (BET analysis versus geometric), the INAS densities for K-feldspar and kaolinite would shift slightly to higher values when using geometric surface areas. Compared to desert dust, soil dust shows a higher ice nucleation activity especially for warmer temperatures (Steinke et al. 2016; Hill et al. 2016). Recent studies showed that soil dust samples featured a significant fraction of organic matter and biological active particles, which might influence the ice nucleation at warmer temperatures (Tobo et al. 2014; Hill et al. 2016).

The high uncertainty of the immersion freezing efficiency of soot aerosol is represented in the deviation of the parameterization line of Murray et al. (2012) and the rescaled Murray-parameterization line for the upper-limiting $n_S$ values from AIDA measurements. Both lines deviate by three orders of magnitude and enclose those for the natural dusts and minerals except of kaolinite.

b. Deposition nucleation

1) DESERT DUST

Figure 4a shows the INAS densities (filled symbols) in the ice saturation ratio–temperature plane for all deposition nucleation experiments on desert dust listed in Table 5. The open symbols show results from two previous AIDA deposition nucleation studies (Möhler et al. 2006; Field et al. 2006). These data points were calculated by dividing the published activated fraction by the aerosol surface area calculated from the median diameter and the aerosol number concentration also given in the papers by Möhler et al. (2006) and Field et al. (2006). The range of the INAS density is indicated by the different colors and typical error bars for temperature and ice saturation ratio are indicated for one data point.

Deposition nucleation experiments with desert dust were so far only conducted at temperatures above 206 K. These experiments yield INAS densities from $5 \times 10^8$ to $5 \times 10^{12}$ m$^{-2}$. The samples AD1, AD2, and SD2 show a similar ice nucleation behavior over the whole temperature range. The samples CID1 and SD19 have a somewhat higher-deposition nucleation activity at lower ice saturation ratios. The INAS density data from ID1 are located between the two other groups.

2) SOOT

Figure 4b shows the INAS densities in the ice saturation ratio–temperature plane for all deposition nucleation experiments with soot listed in Table 6. Crawford et al. (2011) already published the onset ice active fractions for the experiments from the campaigns IN09 and IN11. However, these experiments were reevaluated because of the refined evaluation algorithm. In Fig. 4b, the soot samples are listed with increasing OC mass content. Typical error bars for the temperature and the ice saturation ratio are shown for one data point. Note that the deposition nucleation INAS densities are not just upper-limit values as were reported for soot immersion freezing.

Deposition nucleation was observed for temperatures from 195 to 235 K within INAS densities ranging from $9 \times 10^8$ to $8 \times 10^{13}$ m$^{-2}$. The INAS density data for a higher OC mass content ($\geq 20$ wt%; triangles and diamonds) lie at higher ice saturation ratios than the results for a lower OC mass content (circles and squares), clearly showing that the OC material suppresses the
deposition nucleation activity of the soot particles. This is in agreement with the findings of Möhler et al. (2005a), who also found a decreasing deposition nucleation activity of CAST soot with increasing OC mass content.

3) PARAMETERIZATION AND DISCUSSION

The measured deposition nucleation data for low-OC-content soot aerosol with u-shaped INAS density isolines was fitted with the following equation:

\[ n_S(T, S_i, \text{aerosol}) = \exp\left(\alpha(S_i - 1)^{1/4}\cos[\beta(T - \gamma)]^2\arccot[\kappa(T - \lambda)]/\pi\right) \text{ m}^{-2} \]

\[ \arccot(x) := \frac{\pi}{2} - \arctan(x). \]

The five parameters \( \alpha, \beta, \gamma, \kappa, \) and \( \lambda \) determine the saddle point of the isolines, the steepness of the isolines left and right from the saddle point, and the distance between the isolines. To get the best-fit values, the data points were classified according to their order of magnitude of the INAS density. The classified dataset was then fitted for a constant INAS density only as a function of temperature and extrapolated beyond the data points. The

\[ n_S(T, S_i, \text{aerosol}) = \exp\left(\alpha(S_i - 1)^{1/4}\cos[\beta(T - \gamma)]^2\arccot[\kappa(T - \lambda)]/\pi\right) \text{ m}^{-2} \]

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parameterization is only valid for ice saturation ratios ranging from 1.0 to the homogeneous freezing threshold and water saturation, respectively. The values of the five parameters for soot with an OC mass content of less than or equal 20 wt% are listed in Table 7. This parameterization approach is also used for the dust deposition nucleation dataset, although no AIDA ice nucleation measurements with desert dust were performed so far for temperatures lower than 206 K. The u-shaped isolines for the ice nucleation efficiency of minerals and ATD were found in laboratory studies (Welti et al. 2009; Hiranuma et al. 2014b; Koehler et al. 2010) guiding the shape of the INAS density isolines for desert dust below 206 K. The fit parameter values for desert dust excluding the sample CID1 are listed in Table 7. Figure 5 shows the parameterization for (Fig. 5a) soot and (Fig. 5b) desert dust together with an INAS density isoline of $n_s = 10^{11}$ m$^{-2}$ and the data points within 5 x $10^{10}$ and 5 x $10^{11}$ m$^{-2}$.

Note that INAS densities above about $3 \times 10^{15}$ m$^{-2}$ (assuming spherical particles with diameters of less than 0.01 μm) are not relevant for atmospheric ice nucleation. The extrapolation on the temperature scale is discussed in section 4. The u-shaped form of the INAS density isolines was already suggested by Hoose and Möhler (2012). The authors defined three regimes determining the ice nucleation behavior below water saturation. The three regimes can be identified in the dataset of the soot and are shown as gray boxes in Fig. 4b. In regime A, the isolines decrease with increasing temperature showing that for the same ice activity a higher ice supersaturation is required. Thomson et al. (2015) explained this behavior from theory with the pressure induced rarification of the water vapor, which leads to an increase in ice saturation ratio and therewith to the limitation of ice nucleation. Regime B is a transition regime between A and C and is defined by nearly horizontal isolines, meaning that $n_s$ only depends on the ice saturation ratio. In regime C, the isolines increase very steeply toward water saturation indicating a strong temperature dependence. Therefore, we assume that ice nucleation in regime C is not dominated by the classical

### Table 6. Conditions for deposition nucleation experiments with soot: $n_{s,0}$ is the initial aerosol number concentration measured by the CPC, $s_{m,0}$ is the initial aerosol surface area concentration; and $T_{start}$, $s_{start}$, and $n_{s,0}$ are the temperature, ice saturation ratio, and INAS density at start of ice nucleation, respectively. When two values are listed in the $s_{m,0}$ column, the first value is the smaller mode and the second value is the larger mode of the aerosol surface area lognormal distribution. If ice nucleation starting conditions are not given (asterisk), no heterogeneous ice nucleation was observed. Previously published experiments are labeled with Cr11 (Crawford et al. 2011).

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<td>ICIS07</td>
<td>15</td>
<td>21 Sep 2007</td>
<td>GSG-Ar</td>
<td>425.86</td>
<td>74.64, 59.52</td>
<td>233.6</td>
<td>1.36</td>
<td>1.144 x 10^11</td>
<td>Cr11</td>
</tr>
<tr>
<td>IN09</td>
<td>8</td>
<td>24 Mar 2006</td>
<td>CAST minOC</td>
<td>209.45</td>
<td>70.02</td>
<td>227.0</td>
<td>1.16</td>
<td>1.147 x 10^10</td>
<td>Cr11</td>
</tr>
<tr>
<td>IN09</td>
<td>21</td>
<td>30 Mar 2006</td>
<td>CAST maxOC</td>
<td>290.00</td>
<td>4.18, 37.32</td>
<td>224.3</td>
<td>1.50</td>
<td>3.327 x 10^10</td>
<td>Cr11</td>
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<tr>
<td>IN09</td>
<td>25</td>
<td>31 Mar 2006</td>
<td>GSG-Ar</td>
<td>433.24</td>
<td>206.37</td>
<td>226.5</td>
<td>1.17</td>
<td>5.570 x 10^10</td>
<td>Cr11</td>
</tr>
<tr>
<td>IN11</td>
<td>15</td>
<td>20 Nov 2007</td>
<td>GSG-N2</td>
<td>99.31</td>
<td>33.75</td>
<td>227.1</td>
<td>1.16</td>
<td>2.973 x 10^10</td>
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<tr>
<td>IN11</td>
<td>20</td>
<td>22 Nov 2007</td>
<td>mCAST minOC</td>
<td>1111.52</td>
<td>166.96</td>
<td>225.0</td>
<td>1.33</td>
<td>3.050 x 10^10</td>
<td>Cr11</td>
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<tr>
<td>IN11</td>
<td>26</td>
<td>26 Nov 2007</td>
<td>mCAST maxOC</td>
<td>1173.01</td>
<td>129.68</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>Cr11</td>
</tr>
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<td>RICE01</td>
<td>22</td>
<td>27 Nov 2012</td>
<td>GSG-Ar</td>
<td>246.55</td>
<td>16.29</td>
<td>220.1</td>
<td>1.24</td>
<td>7.311 x 10^10</td>
<td>Cr11</td>
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<tr>
<td>RICE02</td>
<td>8</td>
<td>29 Apr 2013</td>
<td>GSG-Ar</td>
<td>51.75</td>
<td>4.33</td>
<td>219.6</td>
<td>1.26</td>
<td>9.642 x 10^10</td>
<td>Cr11</td>
</tr>
<tr>
<td>RICE02</td>
<td>9</td>
<td>30 Apr 2013</td>
<td>GSG-Ar</td>
<td>94.44</td>
<td>5.61</td>
<td>219.2</td>
<td>1.25</td>
<td>1.801 x 10^12</td>
<td>Cr11</td>
</tr>
<tr>
<td>RICE02</td>
<td>15</td>
<td>2 May 2013</td>
<td>GSG-Ar</td>
<td>48.25</td>
<td>0.89</td>
<td>220.0</td>
<td>1.23</td>
<td>7.968 x 10^11</td>
<td>Cr11</td>
</tr>
<tr>
<td>SOOT11</td>
<td>17</td>
<td>2 Dec 2010</td>
<td>Diesel</td>
<td>4852.92</td>
<td>97.10</td>
<td>230.8</td>
<td>1.43</td>
<td>7.776 x 10^10</td>
<td>Cr11</td>
</tr>
<tr>
<td>SOOT11</td>
<td>21</td>
<td>3 Dec 2010</td>
<td>Diesel</td>
<td>1900.00</td>
<td>39.40</td>
<td>210.74</td>
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<td>Cr11</td>
</tr>
<tr>
<td>SOOT11</td>
<td>24</td>
<td>3 Dec 2010</td>
<td>Diesel w/o pf</td>
<td>110.00</td>
<td>57.04</td>
<td>210.02</td>
<td>1.47</td>
<td>2.079 x 10^10</td>
<td>Cr11</td>
</tr>
</tbody>
</table>

### Table 7. Fit parameters of Eq. (7) for desert dust and soot with an organic carbon content of less than or equal 20 wt%.

<table>
<thead>
<tr>
<th>Aerosol</th>
<th>$\alpha$</th>
<th>$\beta$</th>
<th>$\gamma$</th>
<th>$\kappa$</th>
<th>$\lambda$</th>
<th>$T_{start}$ (K)</th>
<th>Valid $T$ range (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Desert dust</td>
<td>285.692</td>
<td>0.017</td>
<td>256.692</td>
<td>0.080</td>
<td>200.745</td>
<td>[206, 240]</td>
<td></td>
</tr>
<tr>
<td>Soot (OC ≤ 20 wt%)</td>
<td>46.021</td>
<td>0.011</td>
<td>248.560</td>
<td>0.148</td>
<td>237.570</td>
<td>[195, 235]</td>
<td></td>
</tr>
</tbody>
</table>
deposition nucleation mechanism but, rather, influenced by the PCF mechanism as recently suggested by Marcolli (2014). This mechanism was recently demonstrated by Wagner et al. (2016) by AIDA ice nucleation experiments with minerals, desert dust, and soot.

Figure 6 shows a comparison of the immersion freezing parameterization and the deposition nucleation parameterization for desert dust and soot. The linear extrapolation of the parameterization line for immersion freezing on desert dust to temperatures between 238 and 245 K would result in INAS densities that are two orders of magnitude higher than the deposition nucleation isoline close to water saturation. For soot, both parameterizations yield similar INAS density values at water saturation and a temperature of about 238 K. However, the $n_S$ parameterization for soot immersion freezing is only an estimate for the upper limit. For soot, the $n_S$ isolines below water saturation are much steeper than the line for immersion freezing, indicating a change in the nucleation mechanism. Because of the steepness of the isolines and this distinct change in the slope at water saturation, we assume that as part of the PCF mechanism the confined water in the pores and cavities freezes homogeneously. For desert dust the isolines below water saturation are much flatter and show a similar slope as the immersion freezing parameterization line. This indicates that the water in pores and cavities of desert dust particles may freeze heterogeneously.

4) COMPARISON TO OTHER DEPOSITION NUCLEATION PARAMETERIZATIONS

Figure 6 shows a comparison of the new parameterization for desert dust and soot below water saturation with parameterizations from the literature for soil dust (Steinke et al. 2016), ATD (Steinke et al. 2015), and hematite (Hiranuma et al. 2014b) and $n_S$ values derived from measurements with kaolinite (Welti et al. 2009). For clarity, only isolines for INAS densities of $10^{10}\, m^{-2}$ are shown.

Comparing the natural dust and dust components parameterization lines, the desert dust is less ice active below water saturation. The higher ice activity of the soil dust might be explained by some biological or organic materials. Note that the ice nucleation in the temperature range between 230 and 250 K was suggested to be triggered by the PCF mechanism, influenced by the porous structure of the samples. ATD might have more suitable cavities in form of cracks due to the milling during manufacturing. Also the soil dust samples might show a more suitable pore structure than desert dust because of the organic or biological materials on the surface. This may also explain the higher ice nucleation activity of the CID1 sample, which was excluded from the parameterization fit.

The soot shows a higher ice nucleation activity than the desert dust in the temperature range between 215 and 235 K. However, soot is less ice active compared to soil dust, ATD, and the dust components hematite and kaolinite.

One striking feature shown in Fig. 7 is the similar slope of all dust sample isolines, which are much flatter than the soot isolines. This supports the suggestion of the previous section: that the pore water freezes homogeneously for soot and heterogeneously for dusts.

4. Discussion of potential use in models and comparison to other parameterizations

Various types of parameterizations for the heterogeneous ice nucleation ability of aerosols have already
been published. Parameterizations based on CNT (e.g., Hoose et al. 2010; Chen et al. 2008; Barahona 2012; Savre and Ekman 2015) depend on parameters that are difficult to determine. Therefore, the free parameters like contact angle or surface tension differ significantly from study to study. Additionally, the results of these parameterizations do not agree with laboratory results over the whole temperature range (Hoose and Möhler 2012). Especially, the suppression of deposition nucleation for warmer temperatures is not reproduced. Savre and Ekman (2015) recently presented a parameterization based on CNT addressing this issue by introducing a contact-angle distribution function and therewith matching better laboratory observations. However, CNT-based parameterizations using contact angle distributions are complex to implement in models and need generally more computing time, in particular, if the depletion of the most efficient INPs is taken into account.

The second type of parameterizations is empirical. Here, the most commonly used aerosol-specific parameterization framework is that of Phillips et al. (2008, 2013). The framework is based on laboratory as well as field studies and includes also the suppression of deposition nucleation for warmer temperatures. Some of the numerous input parameters that define, for example, the aerosol composition are difficult to be derived from atmospheric

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**Fig. 6.** Comparison of the INAS density parameterization for immersion freezing (solid dark blue line) and deposition nucleation (blue contours) for different water saturation ratios. (a) Desert dust with a linear extrapolation of the immersion freezing line (dashed) and (b) soot.

**Fig. 7.** Comparison of the new deposition nucleation parameterization for desert dust and soot with parameterizations for soil dust, Arizona test dust, and the dust components hematite and kaolinite. For clarity, only isolines for $n_S = 10^{10}$ m$^{-2}$ are shown. Note that all shown parameterizations are only valid for $S_i \approx 1.0$. The black dashed line indicates the ice saturation ratio $S_i$ at water saturation and the gray dotted line the homogeneous freezing threshold for solution droplets ($\Delta a_w = 0.34$; Koop et al. 2000).
measurements (DeMott et al. 2010). More simple descriptions of heterogeneous ice nucleation (e.g., DeMott et al. 2010, 2015) are easier to use in models but, in general, are not applicable to all atmospherically relevant conditions or atmospherically relevant aerosol types. In the case of a Saharan dust layer, DeMott et al. (2015) showed good agreement between their parameterization and the AIDA-based parameterization of Niemand et al. (2012).

Until now, there is no parameterization framework available that is based on well-constrained laboratory experiments covering the whole atmospherically relevant temperature and humidity range. The parameterization framework presented here is in principle limited to the detection limits of the AIDA cloud chamber instruments and the number of evaluated experiments. Therefore, also the AIDA parameterization has to be extrapolated to cover the atmospheric relevant temperature and ice saturation ratio ranges when used in models. In particular, in the cirrus regime, ice nucleation data are lacking at high ice supersaturation and temperatures below 200 K, which makes the parameterization more uncertain in these regions.

The dashed red lines in the top panels of Fig. 8 show the proposed extrapolation of the developed immersion freezing parameterization. The parameterization line is not extrapolated to higher temperatures, because this study as well as previous laboratory studies did not observe ice nucleation of desert dust as well as main dust components above 260 K (Hoos and Möhler 2012). Between 245 K and the homogeneous freezing temperature, the parameterization is linearly extrapolated, because DeMott et al. (2011) showed no leveling off of the ice active fractions from different measurements with Saharan dust. For soot, the rescaled parameterization line from Murray et al. (2012) is simply extrapolated to the homogeneous freezing temperature and to the melting temperature. Below water saturation and temperatures above 240 K, the ice activated fraction is much lower than 1% for a typical aerosol diameter of 1 μm. Therefore, the extrapolation to warmer temperatures is unproblematic (see Fig. 5). However, the extrapolation to higher ice saturation ratios has to be limited to INAS densities less than or equal 1.0 × 10^{15} m^{-2}.

The parameterization framework developed in this study is simple to implement into models. If the dust and soot surface area is provided in size bins, the number of INPs can be calculated via

\[
n_{\text{INP}}(T,S) = \sum_{j} n_{\text{INP},j} \left[1 - \exp\left(-n_{3}S_{\text{dust},j}\right)\right] + \sum_{l} n_{\text{INP},l} \left[1 - \exp\left(-n_{3}S_{\text{soot},l}\right)\right],
\]

where \(j\) is the index of the size bins for dust and \(l\) the index of the size bins for soot; \(n_{\text{INP},j}\) is the number concentration, and \(S_{\text{dust},j}\) and \(S_{\text{soot},l}\) the surface area of dust or soot in size bin \(j\). If only the total surface area is available, this equation can be simplified to

\[
n_{\text{INP}}(T,S) = n_{\text{INP},j} \left[1 - \exp\left(-n_{3}S_{\text{dust},j}\right)\right] + n_{\text{INP},l} \left[1 - \exp\left(-n_{3}S_{\text{soot},l}\right)\right].
\]

The aerosol number concentration and surface area are usually either predicted by interactive aerosol modules in regional or global models (Vogel et al. 2009; Stier et al. 2005) or can be prescribed from climatologies (Hande et al. 2015). To calculate the maximum ice saturation ratio \(S\) reached in models, an algorithm taking into account the competition between homogeneous freezing and heterogeneous nucleation (e.g., Barahona and Nenes 2009; Kärcher et al. 2006) should be used.

Figure 8 shows the new parameterizations for desert dust and soot together with the schemes of Phillips et al. (2013) and Savre and Ekman (2015). For the Phillips parameterization, the equivalent INAS density (Hoose and Möhler 2012) is shown, with a black carbon INN propensity fraction of \(\Xi = 0.1\). The equivalent INAS densities for the Savre and Ekman (2015) parameterizations were recalculated from the published ice active fractions and the median diameters [Eq. (2)]. As the parameterization for dust immersion freezing from Savre and Ekman (2015) is based on the data of Niemand et al. (2012), the parameterization line shows a quite similar behavior compared to the new line. In contrast, the Phillips et al. (2013) parameterization yields much lower INAS densities for \(T < 253\) K. The “leveling off” behavior shown in both schemes is not supported by AIDA laboratory data. For soot immersion freezing, the framework of Phillips et al. (2013) and the new scheme show similar INAS densities, whereas Savre and Ekman (2015) yields INAS densities that are up to three orders of magnitude higher over the whole temperature range. The leveling off for lower temperatures is not shown in the scheme of Savre and Ekman (2015).

For deposition nucleation, the scheme of Savre and Ekman (2015) covers only a small temperature range because of the used datasets [dust: Kanji et al. (2011), soot: Möhler et al. (2005b)]. Therefore, the application of this scheme is doubtful especially for cold cirrus clouds. However, the order of magnitude of the INAS density is comparable to the values of the new scheme. The parameterization of Phillips et al. (2013) does not show the observed u-shaped isolines and shows overall lower INAS density values for a given temperature and ice saturation ratio for both dust and soot. Because of the shape of the isolines, the scheme of Phillips et al. (2013) would give significant INP concentration for soot
only near water saturation. Below about 220 K, the INAS densities from the Phillips parameterization are very small and for ice saturation ratios above 1.6, homogeneous freezing of solution droplets would be more likely.

5. Conclusions

The comprehensive database from AIDA cloud chamber experiments of the last 11 yr was used to develop a parameterization framework for immersion freezing and deposition nucleation. The objective was to parameterize the ice nucleation ability in terms of the INAS density for desert dust and soot but to develop the parameterization in a way extendable to other aerosol types. The already-existing INAS density approach for immersion freezing was extended to deposition nucleation.

As part of the evaluation algorithm, the determination of the aerosol surface area concentration from SMPS and APS measurements was improved by accounting for the reduction of the total surface area concentration due to incomplete CCN activation and by adding a time-bin-wise correction term concerning the reduction of the total aerosol surface area due to the ongoing activation of aerosol particles to ice particles. Therewith, the calculation of the INAS density is more accurate.

For immersion freezing on desert dust, the reanalysis of the AIDA data confirmed the temperature trend of the parameterization line from Niemand et al. (2012) but gave somewhat higher values. The range of variability for INAS densities around the fit line was about the same for samples collected from ground and samples that were transported through the atmosphere, indicating that the fit line may be representative for a wide range of atmospheric desert dust types.

For immersion freezing on desert dust, an exponential function is describing the relation of INAS density and temperature. The comparison of the parameterization
for desert dust with dust components shows that the temperature trend of the INAS density line for illite is closest to our INAS fit line for desert dust. For immersion freezing on soot, only upper-limit INAS densities were determined and fitted by the rescaled parameterization line from Murray et al. (2012). The comparison of the different soot results indicates a high variability for the ice nucleation efficiency of different soot types.

For the ice nucleation activity of desert dust in the cirrus regime, some influence of the sample origin was found. In particular, the sample CID1 shows a higher ability than the other samples, in contrast to the findings in the immersion freezing regime. This difference may be caused by the different porous structure of the aerosol samples, in particular, in regimes where the PCF mechanism dominates the ice nucleation behavior. The soot samples showed a strong influence of the organic carbon content on ice nucleation ability with lower ice nucleation efficiency at higher organic carbon content.

In the deposition nucleation mode, u-shaped isolines of INAS density were found for soot and desert dust. Therewith, the deposition nucleation regime seems to be split into three subregimes. The strong temperature dependency at higher temperatures with INAS density steeply decreasing with temperature my caused by the PCF mechanism. Toward lower temperatures, the $n_s$ isolines turn to negative slope with decreasing temperature, meaning that, at constant $S$, the INAS density increases with temperature increase. There is a transition between both regimes with almost no temperature dependence of the INAS densities. The here-suggested INAS density parameterization allows for the description of the ice nucleation activity in these regimes. For temperatures above 220 K, soot with a low OC content is much more ice active than desert dust, which might be caused by the PCF mechanism. The water confined in pores and cavities is assumed to freeze homogeneously in the case of soot, whereas the water confined in desert dust pores seems to freeze heterogeneously.

For both ice nucleation modes, more experiments are needed to fill gaps in the datasets and to provide a more robust base for accurate parameterizations, in particular at high ice supersaturations and low temperatures of the cold cirrus regime. We also plan to evaluate AIDA experiments with other atmospherically relevant aerosols like organics and coated aerosols in order to extend the INAS density parameterization.

The new parameterization framework covers the most important INP species (dust and soot) in the troposphere (Cziczo et al. 2013). However, the parameterizations are limited to uncoated aerosol particles, although many aerosols are transported over long distances and therewith possibly coated before contributing to ice formation. Möhler et al. (2005b) found that soot coated with sulfuric acid is less ice active in the cirrus temperature regime than uncoated soot. Also an increasing organic carbon content of the soot aerosol reduces its ice nucleation efficiency, but because of only limited data we were not able to quantify this deactivation so far.

In comparison to other parameterization frameworks based on CNT or empirical studies, the here-presented framework has the benefit of a small set of input parameters. Therewith, the framework can more easily be implemented into models. The comparison of the new formulation with a CNT-based and an empirical framework showed large differences in INAS densities especially below water saturation. Hence, when applied in models, the number of ice-nucleating particles resulting from the different parameterizations will significantly differ from each other. Therefore, tests in weather and climate models and constraints to atmospheric measurements of INP concentrations are needed to validate the parameterization schemes and their accuracy for calculating atmospheric INP concentrations as a function of aerosol abundance.

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**APPENDIX A**

**List of Symbols**

- $d$: Aerosol diameter ($\mu$m)
- $d_{act,k}$: INP activation aerosol diameter for time bin $k$ ($\mu$m)
APPENDIX C

Correction of the Aerosol Surface Area Concentration

a. Reduction due to incomplete CCN activation

Both aerosol number and aerosol surface area concentration are lognormally distributed as a function of the aerosol diameter. For some immersion freezing experiment in the AIDA cloud chamber not all aerosol is activated to droplets. Therefore, the total aerosol surface area concentration available for freezing is not equal to the total concentration obtained from the lognormal distribution function. If the CCN activated fraction $f_d$ is less than or equal to 50%, then a correction was done; otherwise, the correction is negligible. The aerosol surface area concentration available for immersion freezing $s_{ae}$ is obtained by integrating the lognormal distribution from zero to the minimum diameter $d_{min}$:

$$s_{ae} = \frac{1}{\sqrt{2\pi}} \int_{0}^{d_{min}} d\log d \frac{s_{ae,0}}{\log d_{g,n}} \exp \left[ -\frac{(\log d - \log d_{m,n})^2}{2\log^2 \sigma_{g,n}} \right].$$  

(C1)

The measured CCN activated fraction $f_d$ is used to calculate the value of $d_{min}$ above which the aerosol was activated to droplets. The fraction of nonactivated aerosol is given by

$$1 - f_d = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{d_{min}} d\log d \frac{1}{\log d_{g,n}} \times \exp \left[ -\frac{(\log d - \log d_{m,n})^2}{2\log^2 \sigma_{g,n}} \right],$$  

(C2)

where $d_{m,n}$ and $\sigma_{g,n}$ are the median diameter and the geometric standard deviation obtained from the fit to the measured size distribution. By substituting the term in the exponential function, the above equation reduces to the standard normal distribution function $\Phi$ with cutoff value $x_{min}$:

$$1 - f_d = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x_{min}} dx \exp \left( -\frac{x^2}{2} \right).$$  

(C3)

Using standard mathematics programs, $x_{min}$ can be determined and therewith the minimum diameter recalculated

$$d_{min} = d_{m,n} \exp(x_{min} \log \sigma_{g,n}).$$  

(C4)
and applied in Eq. (C1). By substituting the term in the exponential function analogously to the number size distribution, the aerosol surface area concentration available for immersion freezing is given by the following equation:

\[ s_{ac} = s_{ac,0} \left[ 1 - \Phi \left( \frac{\log d_{min} - \log d_{m,s}}{\log \sigma_{g,s}} \right) \right], \]  

(C5)

where \( s_{ac,0} \) is the total aerosol surface area concentration obtained from the fit to the measured size distribution and \( d_{m,s} \) and \( \sigma_{g,s} \) the appropriate median diameter and geometric standard deviation, respectively.

\subsection*{b. Correction due to IN activation}

Section 2b described the splitting of the ice nucleation interval in bins \( k \). During each bin \( k \), a certain number of aerosol particles is ice activated and, therefore, is not available for further activations. Hence, the aerosol surface area concentration is reduced in each bin depending on the ice activated fraction. Based on the lognormal size distribution and assuming that the largest particles activate first, the aerosol surface area not activated in bin \( k \) and, therefore, available for bin \( k+1 \) is then given by

\[ s_{ac,k} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\log d_{act,k-1}} \frac{d}{\log \sigma_{g,s}} s_{ac,0} \exp \left[ -\frac{(\log d - \log d_{m,s})^2}{2 \log \sigma_{g,s}} \right], \]

(C6)

where \( s_{ac,0} \) is the total aerosol surface area concentration (if necessary, reduced because of incomplete CCN activation) obtained from the fit to the measured size distribution and \( d_{m,s} \) and \( \sigma_{g,s} \) are the appropriate median diameter and geometric standard deviation, respectively. Similar to the previous section, the activation diameter \( d_{act,k-1} \) is determined from the number size distribution:

\[ 1 - f_{I,k-1} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\log d_{act,k-1}} \frac{d}{\log \sigma_{g,s}} \frac{1}{2 \log \sigma_{g,s}} \exp \left[ -\frac{(\log d - \log d_{m,n})^2}{2 \log \sigma_{g,n}} \right], \]

(C7)

where \( f_{I,k-1} \) is the measured ice activated fraction in bin \( k-1 \), and \( d_{m,n} \) and \( \sigma_{g,n} \) are the respective median diameter and geometric standard deviation obtained from the fit to the measured size distribution. By substituting the term in the exponential function, the integral term again reduces to the standard normal distribution function \( \Phi \) with cutoff value \( x_{act,k-1} \). Using standard mathematics programs, the cutoff value can be determined and \( d_{act,k-1} \) can be recalculated:

\[ d_{act,k-1} = d_{m,n} \exp(x_{act,k-1} \log \sigma_{g,n}). \]

(C8)

Applied in Eq. (C6), the available aerosol surface area concentration in bin \( k \) additionally corrected for dilution is given by

\[ s_{ac,k} = s_{ac,0} \frac{p_k}{p_0} \Phi \left( \frac{\log d_{act,k-1} - \log d_{m,s}}{\log \sigma_{g,s}} \right). \]

(C9)

Note that \( s_{ac,0} \) has to be corrected first owing to incomplete CCN activation as described in the previous section.

\section*{REFERENCES}


