

## Responses to Reviewer 2

First of all, we would like to thank the reviewer for suggestions which help to improve the new version of the manuscript.

Major comments:

*Previously (e.g. Ervens and Feingold, ACP, 2012; also from the same group Ervens and Feingold, GRL, 2013; Kulkarni et al., ACP, 2012, Fan et al., JGR, 2010 and references therein in these papers) also explored the role of temperature, IN size, nucleation schemes on cloud properties. This previous work in the context of present work should be discussed in the section 1 and 4.*

Previous papers as suggested by the reviewer have been included in Section 1. In Section 4, a new sub-section has been added including comparisons of the present results to the outcome of previous model simulations.

*Sensitivity studies are very helpful, but these would be more meaningful if some results are compared against observations (remote sensing for example or laboratory measurements). Constraining against observations would help to understand the influence of microphysical processes, and also dynamics to certain extent. Previous lab data can be plotted in Figure 12a and discussed.*

The new sub-section 4.4 includes also comparisons of present results to atmospheric measurements. A comparison to previous laboratory data as suggested by the reviewer seems not to make much sense as the parameterizations themselves are based on laboratory results. Also we think that Figure 12 is already too busy to add further data.

Minor comments:

*Section 2.3.2: It is not clear how eqn. 16 is used in the model. Is it used after the ice is nucleated? Deposition ice nucleation parameterization gives number of ice particles that meet the  $r^*$  criteria. No additional  $r^*$  tests are required.*

The reviewer is right that the parameterization implicitly includes the  $r^*$  criteria. Nevertheless, it was used before ice nucleation to exclude from the first any calculations with too small particles. This is explained in the revised manuscript.

Section 4.1.3: *Any references that define the strength of weak and strong convection. It is assumed that for weak convection the  $dT$  is 1.5K, but one can argue that it can be classified as a strong convection or mild convection based on the location.*

We agree that the classification of the strength of convection is dependent on the location. In the revised manuscript, the distinction of weak or strong convection has been avoided. We now mention only the used temperature differences (i.e.,  $dT=1.5K$ , 2K, 3K) and the corresponding final temperatures after the ascent of the air parcel (i.e., -24.5, -29, and -40°C).

Section 5: *I suggest clearly mark a paragraph to aid the reader to read the conclusions. Currently, both summary and conclusion are somewhat difficult to separate.*

Following the reviewer's suggestion, this section has been completely rewritten so that the reader can clearly separate summary and conclusions.

Figures 1, 4: *Can you compare to laboratory data.*

The parameterizations given in these figures are directly based on laboratory results. Partly they were derived by the experimentators themselves and included in the references, e.g. immersion freezing by kaolinite particles from Murray et al. (2011), by feldspar particles from Atkinson et al. (2013). The authors decided to avoid showing also all the laboratory data in the figures because this would be too much information in the figures. We would like to avoid additional figures because there are already many tables and figures included. Readers might take a look at the given references to see the data points from the measurements.

In Figure 5 the laboratory data were included because they have not been published yet and the authors had received them by personal communication.

Figure 11: *Any particular reason why immersion + deposition is not considered. Also combination of feldspar and montmorillonite.*

Our intention was to restrict ourselves to some meaningful cases so that the paper would not be too long. The combination contact and immersion mode was selected as both interact with supercooled drops, the combination contact and deposition freezing as both interact with dry particles. So the competition between these freezing modes was investigated. The combination immersion and deposition was studied but not included in the paper in detail.

The combination feldspar (immersion) + montmorillonite (contact) as suggested by the reviewer did not lead to any meaningful results as immersion freezing with feldspar is anyway dominant.

The combination kaolinite (immersion) + feldspar (contact) was selected as here contact freezing was similarly effective as immersion freezing.

Some sentences to explain this have been added to the revised manuscript.

Figure 12: *Is busy plot. If possible simplify it further or divide it.*

The authors agree that this plot is rather busy but we cannot find a possibility to simplify them. To divide the plots would make the comparisons even more difficult. Therefore, the authors decided to leave Figure 12 as it is although the reader has to take some time for it. We have added more detailed explanations in the revised text to help the reader.

### Responses to Reviewer 3

First of all, we would like to thank the reviewer for elaborated suggestions which help to improve the new version of the manuscript.

Major comments:

#### 1. Comparison to previous studies

a) *Several previous studies compared the effect of various freezing modes and other parameters (e.g. IN composition) on mixed-phase cloud properties. Only a few of them are mentioned in the introduction; however, in the result section, the current results are not discussed in the context of previous findings. Such studies include but are not limited to (Fridlind et al., 2007; Eidhammer et al., 2009; Ervens et al., 2011; Kulkarni et al., 2012; Hiron and Flossmann, 2015).*

As suggested by the reviewer, comparisons of the current results to previous ones have been included into a new sub-section 4.4. Comparisons are made to model simulations as well as to observations in real clouds.

b) *How realistic are the assumptions of 1% or 10% of all particles being IN? How are such ratios treated in previous models and justified based on observations?*

In previous models, often fixed numbers of ice nucleating particles were defined (e.g., Ervens et al., 2011) or the parameterization of ice nucleation is not related to particle numbers but empirically related to water samples (e.g., the well-known Bigg parameterization, Bigg, 1953). We did not find similar ratios in previous literature but we used it for our sensitivity studies. The fractions  $F_{INP}$  which are related to the initial aerosol particle number size distribution were compared to observations from field campaigns so that at least it is justified that such fractions of mineral dust are realistic. However, the reviewer is right that not all of these must have the property to act as INP. Furthermore, it is not realistic that all mineral compounds are purely made of, e.g. feldspar. Therefore, we conducted the sensitivity studies to see which fractions are required to form mixed-phase clouds and which INP types are required. So we can conclude that, e.g., feldspar and illite are essential parts of efficient atmospheric ice nucleating particles. Via immersion freezing, mixed-phase clouds were formed already with  $F_{INP}$  of 0.01%. The discussion about these points has been extended in the revised paper.

c) *How do the findings compare to observations? For example, several studies have discussed a likely predominance of immersion and/or contact freezing in the atmosphere (de Boer et al., 2010; Lance et al., 2011 and others).*

Discussions in previous papers as suggested by the reviewer about the predominance of immersion or contact freezing in atmospheric clouds are still open as sampled ice particles do not show an evidence about their freezing process. In particular the role of contact freezing is rather unclear (e.g., Lance et al., 2011). So one can only speculate about the major freezing processes. The current findings may help for this. The discussion in the revised manuscript has been extended. See also comment a) about comparisons to atmospheric clouds.

d) *Your study seems to imply that contact freezing is likely not important. How do these findings agree with previous studies (Ladino et al., 2013, and references therein)?*

In Ladino et al. (2013) findings about contact freezing in laboratory experiments are summarized. In their conclusions the question was listed if contact freezing might be an effective mechanism in real atmospheric cloud situations. Our findings indicate that it might be of minor importance, in particular inside clouds where the amount of inactivated particles is reduced. Another important point is that in our new parameterization the particle size is considered because new experiments validated the conjecture that small particles act as INP at lower temperatures only (e.g., Hoffmann et al., 2013). Therefore, using our previous contact freezing parameterization (Diehl et al., 2006) we most likely overestimated the effects of contact freezing. Some sentences have been added to the revised manuscript to state these points.

## 2. Novelty and uncertainty of results

*Some of the conclusions of the current manuscript are similar to those in studies mentioned in Comment 1. What are new results in the current study? What are the most uncertain and sensitive parameters that affect cloud properties?*

The section Summary and Conclusions has been completely rewritten to underline the new findings of the present simulations.

New findings are that contact freezing is most likely the least important freezing process in spite of the high efficiency of contact ice nucleating particles in laboratory studies. Both contact and deposition freezing require high fractions of potential INP to affect mixed-phase clouds while this happens via immersion freezing already with low INP fractions. Essential components of atmospheric mixed ice nucleating particles are bacteria, feldspar, and illite. The different freezing processes are not in competition with other but they complement each other. The sensitive parameters affecting cloud properties are temperature, aerosol particle composition and concentration, and particle size distribution.

## 3. Mass- vs. surface-based parameterizations

*Several recent laboratory and model studies describe ice nucleation based on surface area of ice nuclei (IN) (Hoose and Möhler, 2012, and references therein). In the current study, some of the data used in the previous study have been converted into mass based parameterizations. I think a surface based parameterization is more intuitive since the number of surface sites determines the IN efficiency. What is the justification of a mass-based parameterization? In particular, in the case of coalesced particles the surface area might change but the mass remains the same which leads then to ambiguous interpretation of the IN activity of the resulting particles. This discrepancy should be discussed and better justified. Assumptions on deposition freezing (p. 16418, l. 16; p. 16430, l. 7) even contradict a surface-based approach. This contradiction should also be better explained.*

During all laboratory experiments which were used as base for the present immersion freezing parameterizations of mineral dust the preparation of the investigated drops was that they contained certain amounts given as mass of the investigated material. Afterwards, from the concentration of the material in the drops, the particle surface area immersed in the drops was calculated. For this purpose the specific surface area (ssa) of the dust particles was used which is strongly dependent on the measurement method. Mostly the BET method is used which is performed by measuring N<sub>2</sub> adsorption isotherm of dry outgassed (i.e., heated in vacuum) powder at 77K. Thus, the value of specific surface area obtained with the non-polar nitrogen substance is used to describe the interaction of the surface with polar water molecules (immersion freezing). This might work well for compact dust particles such as quartz or feldspar but might not work when applied to highly porous aggregates like illite. As it is unknown what part of the inter-crystalline surface is accessible to nitrogen the value of ssa might be afflicted with high errors. Furthermore, using the specific surface area represents just a direct relation between particle mass and surface but not a surface area distribution.

Therefore, and because the original parameter studied in the experiments was the mass of particles in the drops, we decided to use  $n_m$ , the number of active sites per unit mass, for the parameterizations. For Snomax, this was anyway the only parameter which was determined in experiments. An explanation has been added to the revised manuscript.

In case of coalescence, the particle masses of the two drops were summed, i.e. the larger drops contained in general more material and, therefore, froze at warmer temperatures.

However, the authors agree that in principle the particle surface accounts for immersion freezing. A comparison of such two approaches could be investigated in a future study.

p. 16430, l. 7: We have to admit that this sentence mentioned by the reviewer was wrong. It has been replaced by a correct statement that in the contact mode small particles are less efficient

at higher temperatures and less efficient in collisions with drops, i.e. most small particles are available for deposition freezing, at least in the median temperature range.

#### 4. Application of the parcel model

a) *I agree with the authors that a parcel model is a very useful tool to learn about microphysical processes and feedbacks. However, it should be clearly stated which limitations such a model presents. For example, does the presence of growing ice particles feed back on the supersaturation? How realistic are such conditions in the atmosphere when particles might fall out of the parcel?*

Growth and shrinking processes in the air parcel do feed back on the saturation ratio. It is iteratively calculated, see Simmel and Wurzler, 2005. An appropriate remark has been added to the new version of the paper. Limitations of the parcel model because of missing sedimentation are also discussed in the revised paper.

b) *Some of the details of the parcel model should be better explained. I got confused in the introduction when it was stated that 'dry air is mixed into the parcel' (p. 16403, l. 24; p. 16406, l. 14). By definition, an adiabatic air parcel does not have any entrainment of surrounding air. How are these mixing effects described within the model?*

The mentioned sentence has been removed from the introduction as it might confuse readers. However, the currently used model is an adiabatic air parcel models *with entrainment*. This represents a more realistic cloud as it allows (similar as in a real cloud) the exchange of particles, heat, and humidity at the edges of the parcel. The description is given in Simmel et al. (2005) as stated in the paper. All the details were not included in the present paper for reasons of brevity.

The entrainment mixes out activated drops and mixes in aerosol particles which are represented by the drop residuals to ensure particulate mass conservation. The detrainment of a drop quantity  $Q$  (liquid water mass, soluble and total particulate mass, particle number) in size bin  $J$  is determined according to Simmel et al. (2005), with  $R_p$  the size of the air parcel,  $|v|$  the absolute velocity of the parcel:

$$\frac{dQ_J}{dt} = \frac{0.6|v|}{R_p} Q_J$$

c) *On p. 16420, l. 8, precipitation from convective clouds is mentioned. This is confusing, if indeed no precipitation is simulated in the model.*

This phrase has been reformulated in the revised manuscript. Precipitation is mentioned to underline the importance of the ice phase.

*5. Presentation of the figures. Almost none of the figures can be understood solely based on their captions. All figure captions should be improved and details should be explained which freezing mode results etc shown.*

Following the reviewer's suggestion, the figure captions have been carefully reformulated and complemented.

Minor comments:

p. 16406, l. 22: *Why does the ice spectrum have the same bins as the aerosol particle/liquid drop one? Aren't ice particles much larger?*

The bins of the ice particles and the aerosol particles/liquid drops are the same in the model, see Diehl et al., 2006. Indeed the ice particles are larger than the drops but the spectra are the same and include all possible sizes.

p. 16412, l. 25: *Which experiments? The ones by Hoffmann and Kiselev?*

It was meant experiments with polydisperse particle samples. The mentioned sentence has been reformulated.

p. 16413, l. 2ff and Figure 3: *Why can a linear behavior of freezing temperature and particle size be assumed? What is the physical basis for this assumption?*

The linear extrapolation of data points measured for small particle sizes is just a crude assumption. The available data points do not suggest any other behaviour. Physically, the surface of a spherical particle increases quadratically with the radius; however, one does not know if this yields also for the number of active sites on the particle surface and real particles are, of course, not spherical.

p. 16415, l. 13ff: *I am not fond of calling unactivated particles 'dry'. I don't think there is any dry particle in the atmosphere since likely all particles contain some water upon hygroscopic growth. 'Unactivated' might be more appropriate.*

The authors fully agree with the reviewer's comment, in particular inside clouds particles are not really dry. The expression 'inactivated' is used now instead.



p. 16417, l. 22: *I do not understand this. The activated fraction should dependent on the temperature. Please, clarify.*

The measured data points did not indicate a clear dependence on temperature although it should be expected. Therefore, the parameterization was derived in dependence on the ice supersaturation only. Implicitly this contains dependence on the temperature as well as the ice supersaturation increases with decreasing temperature. The mentioned sentence has been reformulated.

p. 16423, l. 2: *I do not see color differences in the tables. Or do you refer here to Figures? (which ones?)*

In the submitted version, the text in Tables 5 to 7 was partly printed in colors to mark mixed-phase and ice clouds, however, this was not possible during publishing in ACPD and, therefore, such cases were marked with bold face. This is mentioned in the figure captions but has not been changed in the text. This has been changed in the revised paper.

p. 16425: *A key reference on bacteria concentrations in clouds is missing here (Delort et al., 2010).*

The mentioned reference has been added.

p. 16428, l. 25: *Do you mean the number of ice particles formed on feldspar and montmorillonite or the total number concentrations of dust particles?*

We meant the number concentrations of ice particles, the sentence has been corrected.

p. 16429, 1<sup>st</sup> paragraph: *Does this discussion refer to results in a figure?*

This discussion still refers to Figure 10; it is mentioned in the revised manuscript.

p. 16431, l. 27: *Thermodynamically, most of the water should be in vapor form. You could simply replace 'total water content' by 'total amount of water in condensed form' (or sth similar).*

The mentioned phrase has been replaced.

p. 16433, l. 16: *I do not understand this discussion. It is true that the concentration of unactivated particles might be larger near cloud edges; however, the (super)saturation is much lower there so that ice nucleation seems less likely.*

This remark refers to contact freezing only where no ice supersaturation is required. The sentence has been reformulated and corrected.

Table 6: *This table seems unnecessary given that there are only a few cells with numbers > 0.01. I think these results could be easily mentioned in the text.*

We would like to keep Table 6 for reasons of completeness.

Figure 1: *Why does the extrapolation of the Snomax result in a plateau?*

As can be seen in Figure 2, this plateau was observed in the measurements of Wex et al. (2015).

Figure 12: *This figure is very busy and not clear. What are the droplets and what are the ice particles?*

The authors agree that this plot is very busy but we cannot find a possibility to simplify them. To divide the plots would make the comparisons even more difficult. Therefore, the authors decided to leave Figure 12 as it is although the reader has to take some time for it. More detailed explanations have been added to the text.

Technical comments:

All technical changes suggested by the reviewer have been changed in the revised manuscript.

Particular comments:

*Tables 2-5: Please, check carefully the equation numbers you refer to here. They seem wrong.*

The equation numbers mentioned with Tables 2 to 5 have been checked and corrected.

*Figure 5 and 6: References to Ardon-Dryer and Levin (2012) and Danielczok and Bingemer (2014) are missing in the reference list.*

According to the technical staff of ACPD, references from personal communication should not be listed in the reference section but mentioned in the text only.

*Figure 11: Clarify the units: Figures say 'per L', caption says 'per cm<sup>3</sup>'.*

The ice particle numbers are given per L, the figure caption has been corrected.

### **List of relevant changes**

- 1) Comparisons of the current results to previous ones have been included into the revised manuscript at several places, in particular in the new sub-section 4.4. Comparisons are made to model simulations as well as to observations in real clouds.
- 2) The section Summary and Conclusions has been completely rewritten to underline the new findings of the present simulations.
- 3) An explanation why the number of active sites per unit mass was used for the parameterization of immersion freezing has been added to the revised manuscript.
- 4) Limitations of the parcel model are discussed in the revised paper.
- 5) The figure captions have been carefully reformulated and complemented.
- 6) More detailed explanations regarding Figure 12 have been added to the text.

# New particle-dependent parameterizations of heterogeneous freezing processes: Sensitivity studies of convective clouds with an air parcel model.

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## 6 **Abstract**

7 Based on the outcome of laboratory results, new particle-dependent parameterizations of het-  
8 erogeneous freezing were derived and used to improve and extend a two-dimensional spectral  
9 microphysics scheme. They include (1) a particle-type dependent parameterization of immer-  
10 sion freezing using the numbers of active sites per mass, (2) a particle-type and size-resolved  
11 parameterization of contact freezing, and (3) a particle-type dependent description of deposi-  
12 tion freezing. The modified microphysical scheme was embedded in an adiabatic air parcel  
13 model with entrainment. Sensitivity studies were performed to simulate convective situations  
14 and the impact of ice nuclei concentrations and types on ice formation. As a central diagnostic  
15 parameter the ice water fraction *IWF* was selected which is the relation of the ice water con-  
16 tent to the total water content. The following parameters were varied: initial aerosol particle  
17 number size distributions, types of ice nucleating particles, final temperature, and the frac-  
18 tions of potential ice nucleating particles. Single and coupled freezing processes were investi-  
19 gated. The results show that immersion freezing seems to be the most efficient process. Con-  
20 tact freezing is constrained by the collision kernel between supercooled drops and potential  
21 ice nucleating particles. The importance of deposition freezing lies in secondary ice for-  
22 mation, i.e. small ice particles produced by deposition nucleation trigger the freezing of su-  
23 percooled drops by collisions. Thus, a broader ice particle spectrum is generated than by im-  
24 mersion and contact freezing. During coupled immersion/contact and contact/deposition  
25 freezing no competition was observed; both processes contribute to cloud ice formation but  
26 do not impede each other. As already suggested in literature, mineral dust particles seem to be

1 the most important ice nucleating particles. Biological particles are probably not involved in  
2 significant ice formation. **The sensitive parameters affecting cloud properties are temperature,**  
3 **aerosol particle composition and concentration, and particle size distribution.**

## 5 **1 Introduction**

6 The importance of the ice phase in mixed-phase convective clouds is indisputable. The addi-  
7 tional release of the latent heat of freezing enforces not only the strength of the convection;  
8 the presence of ice particles in the cloud also substantially modifies the dynamical structure  
9 and the amount of precipitation (e.g., Gilmore et al., 2004). Hence, studying the ice phase in  
10 convective mixed-phase clouds is highly relevant for the understanding of such clouds and  
11 their atmospheric impact. With the convective updraft water drops are transported into re-  
12 gions where the temperature is low enough to allow them to freeze. Homogeneous freezing  
13 (i.e., freezing that does not require the presence of ice nuclei) becomes efficient at tempera-  
14 tures below  $-35^{\circ}\text{C}$  (Pruppacher and Klett, 2010). Thus, at warmer temperatures in the tropo-  
15 sphere heterogeneous freezing (which involves ice nucleating particles) is the only process of  
16 ice initiation, potentially triggering secondary ice formation. Heterogeneous freezing signifi-  
17 cantly changes the availability of liquid water in the upper parts of the cloud since ice parti-  
18 cles grow at the expense of liquid drops by the deposition of water vapor (Bergeron-Findeisen  
19 process) and by riming (i.e., collection of liquid water). Thus, the number of ice nucleating  
20 particles and their efficiency to initiate ice formation at temperatures above the level of ho-  
21 mogeneous freezing determines the nature of convective clouds as it modifies cloud micro-  
22 physical processes and cloud development (e.g., van den Heever et al., 2006; Ekman et al.,  
23 2007; Phillips et al., 2007; Lee et al., 2009).

24 For detailed investigations of cloud microphysical processes adiabatic parcel models with  
25 entrainment are often employed (e.g., Simmel et al., 2005; Leroy et al., 2006; Diehl et al.,  
26 2006; Diehl et al., 2010; Ervens and Feingold, 2012). Air parcel models describe a rising  
27 bubble of air whose volume increases with height. The advantage of parcel models is that  
28 they allow a detailed description of the cloud microphysical processes, usually achieved by  
29 the use of spectral bin-microphysical models that explicitly solve the microphysical equa-  
30 tions (see Khain et al., 2000, for an overview).

1 The initiation of the ice phase in numerical models is always parameterized. As the ability of  
2 atmospheric particles to serve as ice nuclei varies over a wide temperature range for each  
3 freezing process (Pruppacher and Klett, 2010), for particular model simulations parameteri-  
4 zations are required which describe the effects of different types of ice nuclei. **Model investi-**  
5 **gations which compared the effects of different ice nucleation schemes (Fan et al., 2010;**  
6 **Kulkarni et al., 2012; Ervens and Feingold, 2013) imply that cloud properties are sensitive to**  
7 **the surface properties of ice nucleating particles. In those studies particle surface properties**  
8 **were described by the contact angle  $\Theta$ . Other parameterizations were related directly to dif-**  
9 **ferent particle types and model studies** showed that certain aerosol types significantly alter  
10 cloud microphysics (Diehl et al., 2006; Lohmann and Diehl, 2006; Phillips et al., 2008;  
11 Hoose et al., 2008, Storelvmo et al., 2008; Lee et al., 2008). They also allow to simulate the  
12 effects of particular aerosols such as biomass burning particles (Diehl et al., 2007), biological  
13 particles (Phillips et al., 2009), bacteria (Diehl et al., 2010), or mineral dust (DeMott et al.,  
14 2015; Hande et al., 2014).

15 The present investigations are part of the German Science foundation (DFG) research group  
16 INUIT (**I**ce **N**uclei **R**esearch **U**n**I**T) which was established to study heterogeneous ice for-  
17 mation in the atmosphere. In laboratory and field studies the number concentrations, chemical  
18 composition, surface properties and sources of atmospherically relevant ice nuclei are investi-  
19 gated in different freezing modes. As an outcome of these experiments, joint parameteriza-  
20 tions are derived to be fed into a cloud model to simulate mixed-phase cloud microphysics  
21 and to quantify the contribution of ice nuclei particle types and freezing modes. For more de-  
22 tails see the INUIT website: [www.ice-nuclei.de](http://www.ice-nuclei.de).

23 One of the models employed during INUIT is an adiabatic air parcel model with entrainment  
24 and a detailed sectional description of the cloud microphysics. It describes immersion and  
25 contact freezing for various ice nuclei types such as mineral dust, soot, and biological parti-  
26 cles (Diehl and Wurzler, 2004; Diehl et al., 2006). Deposition ice nucleation had not been  
27 included so far as it has been supposed to be of less importance for ice formation in convec-  
28 tive clouds. It might be negligible in mixed phase clouds because water saturation is reached  
29 in the updraft after short times (e.g., Ansmann et al., 2008, based on LIDAR observations).  
30 However, the formation of only a comparably small amount of ice (initiated by few very effi-  
31 cient ice nuclei) might result in secondary ice nucleation with highly complex interactions and  
32 consequences. The present version of the model was improved in the way that now it contains

1 all heterogeneous freezing processes (deposition, contact, coupled condensation/immersion).  
2 This allows to investigate the competition of the freezing modes to understand the importance  
3 of deposition freezing in comparison to immersion and contact freezing. The effects of vari-  
4 ous ice nucleating particles were compared to each other, also considering the different freez-  
5 ing modes, to estimate their importance.

## 6 **2 Model description**

7 The impact of ice nucleating particles on mixed-phase convective clouds is investigated with  
8 a microphysical scheme embedded in an air parcel model with entrainment. The previous ver-  
9 sion of Diehl et al. (2006) was further developed in the way that new parameterizations were  
10 added or existing ones were modified or completely replaced.

11 The model of Diehl et al. (2006) contains a two-dimensional spectral microphysics scheme  
12 which divides the hydrometeor spectra into size bins (Simmel and Wurzler, 2006). These de-  
13 scribe the number and mass of the drops or ice particles within the corresponding size range.  
14 A fixed bin structure is used combining the wetted aerosol particles and the drops in one spec-  
15 trum where the soluble and total mass of aerosol particles is explicitly considered in every  
16 bin. An initial dry aerosol particle number size distribution is given where the particles are  
17 internally mixed with variable insoluble and soluble fractions. After starting the rise of the air  
18 parcel, the particles grow into the droplet part of the spectrum by condensation. The size spec-  
19 tra are allowed to evolve freely, they are not constrained by an underlying distribution func-  
20 tion.

21 Two-dimensional means here that the microphysics is not a function of the drop size only  
22 (one-dimensional case) but a function of both drop and aerosol particle size (Simmel and  
23 Wurzler, 2006). In the one-dimensional case, it is affected that equal sized drops contain only  
24 equal sized particles which affects that drops of the same sizes freeze at the same temperature  
25 (Diehl and Wurzler, 2004). However, in real clouds equal sized drops contain different sized  
26 particles which affect the freezing temperature of the drops. The soluble parts of the aerosol  
27 particles might lead to a freezing point depression (Koop et al., 2000; Diehl and Wurzler,  
28 2004) while the insoluble fractions might give higher freezing temperatures via immersion  
29 freezing (see Section 2.1). Thus, the two-dimensional description of the microphysics allows  
30 that drops of the same sizes freeze at different temperatures which reflects drop freezing in  
31 atmospheric clouds in a more realistic way. It is divided into 90 categories for the particulate  
32 mass and into 66 categories for the water mass, both starting with  $0.002 \mu\text{m}$  in diameter, with

1 a mass doubling in every category for the water mass and a mass doubling in every other cat-  
2 egory for the particulate mass. This combination is recommended in Simmel and Wurzler  
3 (2006).

4 The warm microphysical processes include growth of particles and drops by water vapour  
5 deposition, shrinking of particles and drops by evaporation, collision and coalescence, and  
6 impaction scavenging of particles. The entrainment of aerosol particles, drops, ice particles,  
7 temperature, and humidity is embedded (Simmel et al., 2005). The cold cloud microphysics  
8 describes immersion and contact freezing in parameterized form for various particle types  
9 (Diehl and Wurzler, 2004; Diehl et al., 2006). Condensation freezing is included implicitly in  
10 immersion freezing. Drops which are nucleated during the ascent of the air parcel by aerosol  
11 particles entrained above the freezing level could freeze immediately by immersion freezing.  
12 The growth of ice particles by water vapour diffusion and by riming (collision with super-  
13 cooled droplets) is considered. Ice particles are sampled in a second spectrum which shows  
14 the same bins as the aerosol particle/liquid drop spectrum. Once drops are frozen or particles  
15 are involved in contact or deposition freezing (see next sections) they are shifted to the ice  
16 particle spectrum. **The saturation ratio during growth and shrinking processes is iteratively**  
17 **calculated (Simmel and Wurzler, 2005).**

18 Collision processes are described by the linear discrete method (Simmel et al., 2002) includ-  
19 ing the collision kernel of Kerkweg et al. (2003). By using the corresponding densities and  
20 terminal velocities, the collision kernel is appropriate for all collision processes between aero-  
21 sol particles, drops, and ice particles such as growth of drops by collision/coalescence, impac-  
22 tion scavenging of particles by drops, contact freezing of supercooled drops, growth of ice  
23 particles by riming, and secondary ice formation., i.e. freezing of supercooled drops by colli-  
24 sion with an ice germ.

25 Because of the explicit descriptions mentioned above the microphysical scheme is a useful  
26 tool to study in detail the link between aerosol particles and the evolution of cloud properties.  
27 The incorporation into an air parcel model has the advantage that all changes in the micro-  
28 physical evolution of the cloud can be attributed to microphysical processes. The model im-  
29 provements presented in the next sections include the following:

- 30 1. An updated particle-type dependent description of immersion freezing which is now re-  
31 lated to the mass of insoluble particles contained in drops,



- 1 2. a modified description of contact freezing which is not only dependent on particle type
- 2 but also particle size-resolved,
- 3 3. a new particle-type dependent description of deposition freezing.

4 All parameterizations are directly based on previous and new laboratory measurements as  
5 described in the following sections.

## 6 **2.1 Immersion freezing**

7 The previous description of immersion freezing (Diehl and Wurzler, 2004) gave the freezing  
8 rate of pure water drops containing ice nucleating particles as a function of the drop volume  
9 according to:

$$10 \quad -\frac{dN_f}{dt} = N_{liq} B_{imm} V_d \exp(-a_{1,imm} T) \frac{dT}{dt} \quad (1)$$

11 with  $N_f$  the number of frozen drops,  $N_{liq}$  the number of liquid drops, and the constants  $a_{1,imm}$   
12 and  $B_{imm}$ . This parameterization implicitly reflected the fact that larger drops contain more  
13 particles because of collision and coalescence of drops and impaction scavenging of aerosol  
14 particles. The previous version was replaced by a new one which is coupled directly to the  
15 mass of insoluble particles in the drops. This is possible because of the sectional distribution  
16 of drops and particles into size classes (Diehl et al., 2006).

### 17 **2.1.1 Parameterizations based on laboratory data**

18 The experimental data used as basis for the parameterizations include the following ice nucle-  
19 ating particle types: bacteria, pollen, feldspar, illite, and kaolinite. Murray et al. (2011) inves-  
20 tigated kaolinite KGa-1b, Atkinson et al. (2013) K-feldspar. Illite NX was studied by Broad-  
21 ley et al. (2012) and recently by Hiranuma et al. (2015). The latter publication summarized  
22 the results from seventeen experimental techniques. Wex et al. (2015) includes the results  
23 from seven experimental techniques using Snomax<sup>®</sup> as a proxy for bacteria. Previous data for  
24 pollen (Diehl et al., 2002; v. Blohn et al., 2005) were newly evaluated. **As parameter for im-**  
25 **mersion freezing the number of active sites per unit mass  $n_m$  was selected as this is derived**  
26 **directly from the particle mass concentration in the drops during the experiments. Some refer-**  
27 **ences give as outcome from their measurements the surface density of active sites per unit**  
28 **particle surface  $n_s$ ; however, this parameter was derived afterwards by using a specific parti-**  
29 **cle surface.**

1 For kaolinite KGa-1b, K-feldspar, tree and grass pollen, an exponential increase of  $n_m$  with  
 2 temperature  $T$  was found which is described by

$$3 \quad n_m = \exp(a_{imm} + b_{imm} T_s) \quad (2)$$

4 with  $n_m$  in  $\text{g}^{-1}$ ,  $a_{imm}$  and  $b_{imm}$  particle-related constants,  $T_s = T_0 - T$ ,  $T_0 = 0^\circ\text{C}$ , with  $T$  in  $^\circ\text{C}$ . The  
 5 constants for all particle types are listed in Table 1. Given in Table 1 are also the parameters  
 6  $T_{ini}$  and  $T_{lim}$ , representing the onset of immersion freezing during experiments and the lowest  
 7 temperature which was investigated in the experiments, respectively.

8 Based on the best fit of the data of Murray et al. (2011) the constants in Eq. 2 were derived for  
 9 kaolinite KGa-1b by using an average specific particle surface area of  $11.8 \text{ m}^2 \text{ g}^{-1}$  as given by  
 10 Murray et al. (2011); the result is shown in Figure 1 as orange line. The solid part of the line  
 11 represents the range which is validated by measurements of Murray et al. (2011) while the  
 12 dotted part shows an extrapolation towards higher temperatures. The value of  $T_{ini}$  is based on  
 13 earlier measurements of Pitter and Pruppacher (1973). The same was performed for K-  
 14 feldspar by using the best fit to the experimental data given by Atkinson et al. (2013) which  
 15 was changed into Eq. 2 by using the specific particle surface area of  $3.2 \text{ m}^2 \text{ g}^{-1}$  from Atkinson  
 16 et al. (2013). The parameters are listed in Table 1, the result of Eq. 2 is shown in Figure 1 as  
 17 red line. Regarding pollen, previous data from Diehl et al. (2002) and v. Blohn et al. (2005)  
 18 were evaluated. From the frozen fractions of drops, the drop volume, and the mass of pollen  
 19 in the drops the numbers of active sites  $n_m$  as functions of temperature were calculated accord-  
 20 ing to (e.g., Murray et al., 2011):

$$21 \quad n_m = -\frac{\ln(1 - f_{ice}(T))}{c_{pollen} V_{drop}} \quad (3)$$

22 where  $f_{ice}(T)$  is the fraction of frozen drop at temperature  $T$ ,  $c_{pollen}$  the pollen concentration per  
 23 drop, and  $V_{drop}$  the drop volume. Data for tree and grass pollen were summarized leading to  
 24 two parameterizations for tree and grass pollen as given in Eq. 2. The constants are listed in  
 25 Table 1 and the results of Eq. 2 are shown in Figure 1 as light green lines. The solid parts of  
 26 the lines represent the ranges which are validated by measurements while the dotted parts  
 27 show extrapolations towards lower temperatures.

28 Regarding illite NX and Snomax<sup>®</sup>, parameterizations were suggested by Broadley et al.  
 29 (2012), Hiranuma et al. (2015), and Wex et al. (2015). To come to analogue descriptions as in  
 30 the cases of the other particle types, these parameterizations were replaced by new ones fol-

1 lowing Eq. 2. Regarding illite NX, the parameterization was derived from the data reported by  
 2 Hiranuma et al. (2015). The fit as given by Hiranuma et al. (2015) was changed into an ex-  
 3 pression for  $n_m$  by using the specific particle surface area of  $124.4 \text{ m}^2 \text{ g}^{-1}$  of the illite NX sam-  
 4 ple (Hiranuma et al., 2015). From the non-linear curve (solid blue line in Figure 2) a linear  
 5 regression line was derived which is included in Figure 1 (blue solid line). For Snomax<sup>®</sup>, it  
 6 was concluded in Wex et al. (2015) that the average  $n_m$  values (given in Figure 2 as open  
 7 green symbols) are very well represented by the Hartmann et al. (2012) parameterization (sol-  
 8 id green line in Figure 2). One can notice a linear increase in the temperature range down to  
 9 approximately  $-9^\circ\text{C}$  and a further progress on a maximum value of  $1.4 \times 10^{12} \text{ g}^{-1}$ . Thus, a lin-  
 10 ear regression curve was derived for the increase of  $n_m$  until the maximum value was reached;  
 11 it is included in Figure 2 as solid green line.

12 It can be noted from Figure 1 that bacteria act at the highest temperatures starting not far be-  
 13 low  $0^\circ\text{C}$ . Pollen lie in the range of the mineral dust particles. The differences between the  
 14 mineral dust types are significant with feldspar the most efficient one, kaolinite the least effi-  
 15 cient one.

### 16 **2.1.2 Treatment of immersion freezing in the model**

17 The previous Eq. 1 which gives the freezing rate of the supercooled drops had to be replaced  
 18 by a similar expression which couples the number of active sites and the mass of insoluble  
 19 particles in the drops. According to the singular description of heterogeneous freezing (Vali,  
 20 1971; Broadley et al., 2012) the frozen fraction of drops  $f_{ice}$  is given by

$$21 \quad f_{ice}(T) = \frac{N_f(T)}{N_{liq}} = 1 - \exp(-n_m(T)m_{pid}) \quad (4)$$

22 with  $N_f(T)$  the number of frozen drops at temperature  $T$ ,  $N_{liq}$  the number of liquid drops,  $m_{pid}$   
 23 the mass of particles immersed in the drops, and  $n_m(T)$  the number of active sites per unit  
 24 mass at temperature  $T$  which is related to the cumulative nucleus spectrum  $K(T)$  per unit  
 25 mass per unit temperature:

$$26 \quad n_m(T) = - \int_{T_0}^T K(T) dT \quad (5)$$

1 when lowering the temperature from  $T_0 = 0^\circ\text{C}$  to  $T$ . From Eqs. 4 and 5 an expression for the  
 2 change of the number of frozen drops  $\Delta N_f$  per temperature interval  $\Delta T$  can be derived (Con-  
 3 nolly et al. (2009) :

$$4 \quad \Delta N_f = N_{liq} (1 - \exp(-K(T) m_{pid} \Delta T)) \quad (6)$$

5 with  $N_{liq}$  the number of supercooled liquid drops and  $m_{pid}$  the mass of particles immersed in  
 6 the drop. Thus, Eq. 1 can be replaced by

$$7 \quad \frac{dN_f}{dt} = N_{liq} \frac{1 - \exp(-K(T) m_{pid} dT)}{dT} \quad (7)$$

8 As the aerosol particles are internally mixed one can assume that only a fraction of the insolu-  
 9 ble mass per drop consists of ice nucleating material. Thus, the mass  $m_{pid}$  was reduced by a  
 10 factor  $F_{INP}$  so that only this mass fraction accounts for possible numbers of active sites  $n_m$  and  
 11 Eq. 7 was modified to

$$12 \quad \frac{dN_f}{dt} = N_{liq} \frac{1 - \exp(-K(T) m_{pid} F_{INP} dT)}{dT} \quad (8)$$

13 A similar treatment was actualized in Diehl and Wurzler (2010). From Eqs. 2 and 5 it fol-  
 14 lows:

$$15 \quad K(T) = \frac{dn_m(T)}{dT} = b_{imm} \exp(a_{imm} + b_{imm} T_s) \quad (9)$$

16 In Eq. 9 the freezing point depression due to the content of soluble material in the drops is  
 17 considered, for details see Diehl and Wurzler (2004). In the model simulations, immersion  
 18 freezing starts at the particle-related temperature  $T_{ini}$ , and at temperatures below  $T_{lim}$  it is as-  
 19 sumed that the numbers of active sites stay constant. The sizes of possibly ice nucleating par-  
 20 ticles are restricted, i.e., dust particles must be larger than  $0.1 \mu\text{m}$  in diameter, bacteria are  
 21 limited to their typical diameters of  $0.3$  to  $2 \mu\text{m}$  (Matthias-Maser and Jaenicke, 1995). Pollen  
 22 are large particles of  $10 \mu\text{m}$  at least (Straka, 1975), however, for the present simulations a  
 23 lower limit of  $2 \mu\text{m}$  was selected to allow at least some freezing by pollen (see Section 3, ini-  
 24 tial dry particle number size distributions).

25 During the model simulations the content of insoluble particles per drop varies by several  
 26 orders of magnitude which is due to the sizes of the condensation particles, the uptake of par-  
 27 ticles by the drops via impaction scavenging, and the drop collisions followed by coalescence.

1 Thus, not all drops of the same sizes freeze at certain temperatures which describes the situa-  
2 tion in real clouds (Diehl and Wurzler, 2004).

## 3 **2.2 Contact freezing**

4 In Diehl et al. (2006) contact freezing was described for several particle types independent on  
5 their sizes. However, measurements indicate that the size of the involved particles affects the  
6 contact freezing efficiency in the way that efficiency increases with increasing particle size  
7 (e.g., Gorbunov et al. (2001); Hoffmann et al., 2013a; Ladino et al. 2013). Therefore, the de-  
8 scription of Diehl et al. (2006) was modified in the way that it is now particle-size dependent.

### 9 **2.2.1 Parameterizations based on laboratory data**

10 Measurements which give a direct correlation between particle size and contact freezing effi-  
11 ciency are not yet available for a wider range of ice nucleating particle types. Therefore, for  
12 the present parameterization different size classes were examined. Hoffmann et al. (2013a,  
13 2013b), and Hoffmann and Kiselev (2014) performed contact freezing experiments using an  
14 electrodynamical balance with monodisperse particles of 150 to 750 nm diameter. The results  
15 showed rather low median freezing temperatures  $T_{50}$  (the temperature where 50% of an ob-  
16 served drop population freeze), e.g.,  $-34^{\circ}\text{C}$  for illite NX,  $-32.5^{\circ}\text{C}$  for kaolinite Fluka and less  
17 than  $-25^{\circ}\text{C}$  for Snomax<sup>®</sup>, in comparison to measurements with polydisperse particle samples.

18 Those experiments with polydisperse particle samples were performed at the UCLA vertical  
19 wind tunnel by Levin and Yankofsky (1983) and Pitter and Pruppacher (1973). The latter  
20 studied kaolinite and montmorillonite with particle diameters between 0.1 and 10  $\mu\text{m}$  with a  
21 mode between 1 and 2  $\mu\text{m}$  and measured median freezing temperatures of  $-12^{\circ}\text{C}$  and  $-8^{\circ}\text{C}$ ,  
22 respectively. Levin and Yankofsky found a median freezing temperature of  $-4.5^{\circ}\text{C}$  for bacte-  
23 ria. Diehl et al. (2012) investigated polydisperse mineral particles with supercooled drops  
24 suspended in an acoustic levitator. Median freezing temperatures were  $-11.5^{\circ}\text{C}$  for illite NX  
25 and  $-8.7^{\circ}\text{C}$  for montmorillonite K10. The latter agrees very well with the value found by Pit-  
26 ter and Pruppacher (1973) within the measurement error (1K). Extrapolating the data of  
27 Hoffmann et al. (2013b) for illite NX towards larger particle sizes as shown in Figure 3 indi-  
28 cates that a median freezing temperature of  $-11.5^{\circ}\text{C}$  dust could be affected by particles with  
29 sizes between 2.3 and 3.0  $\mu\text{m}$  in diameter (Diehl et al., 2012). Those particles are part of the  
30 polydisperse particle spectrum of illite NX (Hiranuma et al., 2015). Thus, one might conclude  
31 that the median freezing temperatures determined for polydisperse particle samples are affect-

1 ed by larger particles present in the size spectrum. This is probably the case also for previous  
2 findings measured with polydisperse particle samples.

3 Therefore, as an approximation, data obtained from polydisperse particle samples were used  
4 in the present parameterizations for particles with diameters mostly larger than 1  $\mu\text{m}$ , at least  
5 larger than 0.7  $\mu\text{m}$ . Data obtained from monodisperse particle samples were taken for ranges  
6 around the respective particle sizes.

7 The experimental data used as basis for the parameterizations include the following ice nucle-  
8 ating particle types: bacteria, feldspar, montmorillonite, illite, and kaolinite. In most cases, a  
9 linear correlation between the frozen fraction of drops and the temperature was found. From  
10 the experimental data, regression lines were calculated which are shown in Figure 4 for vari-  
11 ous particle types (marked by different colors) and particle sizes (marked by different line  
12 styles).

### 13 Bacteria

14 Particle sizes of bacteria were restricted to their typical sizes with diameters between 0.3 and  
15 2  $\mu\text{m}$  (Matthias-Maser and Jaenicke, 1995). Measurements of Hoffmann and Kiselev (2104)  
16 were performed with monodisperse Snomax<sup>®</sup> particles of 0.32 and 0.55  $\mu\text{m}$  diameter. These  
17 data were used for size ranges from 0.3 to 0.5  $\mu\text{m}$  and from 0.5 to 0.7  $\mu\text{m}$ . For particles be-  
18 tween 0.7 and 2  $\mu\text{m}$  the data of Levin and Yankofsky were taken. The results are given in  
19 Figure 4 as green lines.

### 20 Mineral dust particles

21 For mineral dust particles, a lower size limit of 0.1  $\mu\text{m}$  in diameter was assumed. Illite NX  
22 particles were investigated by Hoffmann et al. (2013b) with 0.15, 0.32, 0.55, and 0.75  $\mu\text{m}$   
23 particles. These data were taken for size ranges from 0.1 to 0.2  $\mu\text{m}$ , 0.2 to 0.4  $\mu\text{m}$ , 0.4 to 0.6  
24  $\mu\text{m}$ , and 0.6 to 0.8  $\mu\text{m}$ . For larger particles, data from Diehl et al. (2012) were used. The re-  
25 sults are shown in Figure 4 as blue lines. K-feldspar was investigated by Hoffmann and  
26 Kiselev (2014) with 0.32 and 0.55  $\mu\text{m}$  particles and by Diehl and Mitra (2014) with polydis-  
27 perse particles. Here three size ranges were defined, from 0.1 to 0.4  $\mu\text{m}$ , 0.4 to 0.8  $\mu\text{m}$ , and  
28 larger than 0.8  $\mu\text{m}$ . The data are marked in Figure 4 as red lines. For kaolinite and montmoril-  
29 lonite, also two size ranges were specified, from 0.1 and 1  $\mu\text{m}$  and larger than 1  $\mu\text{m}$ . The data  
30 for the larger particles sizes were both taken from Pitter and Pruppacher (1973, polydisperse  
31 particle samples). Results from Hoffmann et al. (2013a) were used for the smaller size range

1 of kaolinite. Under the assumption that the differences between larger and smaller INP are  
2 similar for kaolinite and montmorillonite the data for the smaller size range of montmorillo-  
3 nite were obtained by a parallel shifting analogue to kaolinite. In Figure 4, results for kaolinite  
4 are given in orange, results for montmorillonite in cyan.

5 From Figure 4 it can be noted that particles in the larger size ranges affect freezing already at  
6 temperatures around  $-10^{\circ}\text{C}$  while smaller particles become active in a temperature range  
7 around  $-25^{\circ}\text{C}$ . Bacteria act at the highest temperatures, kaolinite and illite at the lowest. For  
8 all particle types and sizes except bacteria smaller than  $0.7\ \mu\text{m}$  the frozen fraction of drops  
9 increases linearly with temperature  $T$  (given in  $^{\circ}\text{C}$ ) according to Diehl et al. (2006):

$$10 \quad \frac{N_f}{N_{liq}} = a_{con} T + b_{con} \quad (10)$$

11 with  $N_f$  the number of frozen drops,  $N_{liq}$  the number of liquid drops colliding with inactivated  
12 particles at temperature  $T$ , and the constants  $a_{con}$  and  $b_{con}$ . Note that in Eq. 10 the frozen frac-  
13 tion is limited to values between 0 and 1. In Diehl et al. (2006), the constants were given for  
14 several particle types independent of their sizes while in the present parameterization the con-  
15 stants  $a_{con}$  and  $b_{con}$  are size-resolved. They are listed in Tables 2 and 3. In case of small bacte-  
16 ria the equation to calculate the frozen fraction of drops with respect to the temperature  $T$  (in  
17  $^{\circ}\text{C}$ ) has the form

$$18 \quad \frac{N_f}{N_{liq}} = a_{con} + b_{1,con}T + b_{2,con}T^2 + b_{3,con}T^3 \quad (11)$$

19 The size-resolved constants  $a_{con}$ ,  $b_{1,con}$ ,  $b_{2,con}$ , and  $b_{3,con}$  are given in Table 2.

## 20 **2.2.2 Treatment of contact freezing in the model**

21 The description of contact freezing includes the following conditions: (1) inactivated particles  
22 have to be present, and (2) particles and supercooled drops have to collide with each other.  
23 Furthermore, the sizes of the particles allowed as activating ice nucleating particles are re-  
24 stricted as for deposition freezing (i.e., dust particles  $> 0.1\ \mu\text{m}$  in diameter, bacteria  $0.3$  to  $2$   
25  $\mu\text{m}$  in diameter).

26 The presence of inactivated particles is always the case during the air parcel ascent because of  
27 entrainment, i.e. new inactivated particles are continuously mixed in at the edges of the simu-  
28 lated cloud. However, in the presently employed air parcel model the particles are in equilib-

1 rium with respect to the water vapor in their environment and, thus, they take up some water  
 2 due to their size and soluble fraction. As introduced in Diehl et al. (2006) the dryness of a  
 3 potential ice nucleating particle is defined by the assumption that the water mass should be  
 4 smaller than half of the dry particle mass.

5 The second condition is considered by a collision kernel  $K$  calculated for supercooled drops  
 6 and particles (Kerkweg et al., 2003; for more details see Diehl et al., 2006):

$$7 \quad K = E_{coll} |V_{\infty,drop} - V_{\infty,ap}| \cdot \pi (r_{drop} - r_{\infty})^2 \quad (12)$$

8 where  $V_{\infty,drop}$  and  $V_{\infty,ap}$  are the terminal velocities of the drop and the particle, respectively,  
 9  $r_{drop}$  and  $r_{ap}$  the drop and particle radii, respectively. The collision kernel shows highest values  
 10 for collisions between large drops and particles (Diehl et al., 2006), i.e. contact freezing is the  
 11 most efficient when large supercooled drops and particles are present. If during the model  
 12 simulations inactivated particles collide with supercooled drops the number of frozen drops is  
 13 calculated. It is assumed that only a fraction  $F_{INP}$  of the aerosol particles is able to act as ice  
 14 nucleating particles. Only drops which collide with those INP are allowed to freeze which is  
 15 included in the following modified Eqs. 13 and 14:

$$16 \quad N_f = F_{INP} N_{liq} (a_{con} T + b_{con}) \quad (13)$$

$$17 \quad N_f = F_{INP} N_{liq} (a_{con} + b_{1,con} T + b_{2,con} T^2 + b_{3,con} T^3) \quad (14)$$

18 This is under the requirement that these equations are based on measurements with one drop–  
 19 particle collision per freezing event so that the fraction of frozen drops in Eqs. 13 and 14 can  
 20 be set equal to the freezing probability (Ladino et al., 2013). This requirement is fully  
 21 achieved in the experiments of Hoffmann et al. (2013a; 2013b), and Hoffmann and Kiselev  
 22 (2014). During the experiments of Pitter and Pruppacher (1973), Levin and Yankofsky  
 23 (1983), and Diehl et al. (2012) the number of collisions per freezing event is not documented.  
 24 Single supercooled drops were freely levitated (in a wind tunnel or an acoustic levitator)  
 25 while one burst of INP was blown on it. Therefore, as the particles collided almost simultane-  
 26 ously with the supercooled drop one could assume that in case the drop froze this was trig-  
 27 gered by the first collision.

## 28 **2.3 Deposition freezing**

### 29 **2.3.1 Parameterizations based on laboratory data**



1 The experimental data used as basis include the following ice nucleating particle types: bacte-  
2 ria, feldspar, illite, Saharan and Asian dust. The measurements were performed with INP  
3 counters FRIDGE (**FR**ankfurt **I**mmersion and **D**eposition freezin**G** **E**xperiment) or with a  
4 continuous flow diffusion chamber (CFDC). Data for Asian and Saharan dust were taken  
5 from measurements with the NAUA FRIDGE (Ardon-Dryer and Levin, 2012), data for illite  
6 NX and Snomax<sup>®</sup> from INUIT FRIDGE experiments (Hiranuma et al., 2015; Danielczok and  
7 Bingemer, 2014; Weber, 2014), and data for illite IMt1 and K-feldspar from CFDC measure-  
8 ments (Yakobi-Hancock et al., 2013). The Snomax<sup>®</sup> data were considered as representative  
9 for bacteria in the model simulations.

10 Ardon-Dryer and Levin (2012) measured the activation of Saharan and Asian dust particles at  
11 different ice supersaturations and temperatures and observed an increase of the activated par-  
12 ticles with supersaturation but no temperature dependence in the observed temperature range  
13 between -15 and -20°C. Figure 5 shows the data of Ardon-Dryer and Levin (2012) as activat-  
14 ed fraction of particles as a function of ice supersaturation. From all data, mean values were  
15 calculated and regression lines were derived. These are included into Figure 6 as cyan and  
16 pink solid lines.

17 The INUIT FRIDGE measurements were performed with illite NX and Snomax<sup>®</sup> in a temper-  
18 ature range from -10 to -25°C. **A significant temperature dependence was not observed and,**  
19 **therefore, the activated particle fraction was derived as a function of ice supersaturation only.**  
20 **As ice saturation is dependent on temperature an implicit dependence of deposition freezing**  
21 **on temperature is already incorporated.** Regression lines were calculated based on the average  
22 values. They are given in Figure 6 as blue (illite NX) and green (Snomax<sup>®</sup>) solid lines. It can  
23 be noticed that illite NX is much more efficient than the Saharan and Asian dust particles  
24 which are characterized by a mixed composition. Asian dust mostly consists of quartz (Möh-  
25 ler et al., 2006) which acts in the deposition mode at lower temperatures than illite (Zimmer-  
26 mann et al., 2008). Also Saharan dust contains a quartz fraction of nearly one third (Möhler et  
27 al., 2006). As expected the activated fractions of Snomax<sup>®</sup> particles are the highest. Meas-  
28 urements with a CFDC were performed at -40°C with illite IMt1 and K-feldspar particles  
29 (Yakobi-Hancock et al., 2013). Regression lines were derived from the data and are given in  
30 Figure 6 as blue (illite IMt1) and red (K-feldspar) broken lines, respectively.

31 For illite NX and Snomax<sup>®</sup>, data are available not only for polydisperse particle samples but  
32 also for samples with distinct particle sizes (Danielczok and Bingemer, 2014; Weber, 2014).

1 In case of illite, monodisperse particles with diameters of 300 and 500 nm resulted in higher  
2 activated fractions with the larger particle sizes but in both cases the activated fractions were  
3 higher than in case of the polydisperse particle samples. In case of Snomax<sup>®</sup>, monodisperse  
4 particles of 100, 200, 300, and 500 nm were investigated finding again higher activated frac-  
5 tions for the larger particle sizes than in the polydisperse case. As particle-size resolved  
6 measurements are not available for all particle types it was decided to treat deposition freez-  
7 ing size-independent in the model. As the activated fractions of the polydisperse particle sam-  
8 ples (which are the basis of the present parameterizations) are lower than the ones of mono-  
9 disperse samples, this would not lead to an overestimation of deposition freezing.

10 For all particle types, an exponential increase of the activated fraction with the ice supersatu-  
11 ration was found which is described by

$$12 \quad \frac{N_{act}}{N_{total}} = \exp(a_{dep} + b_{dep} S_{ice}) \quad (15)$$

13 with  $N_{act}$  the number of activated particles,  $N_{total}$  the total particle number,  $S_{ice}$  the ice super-  
14 saturation given in %,  $a_{dep}$  and  $b_{dep}$  particle-related constants. The values of  $a_{dep}$  and  $b_{dep}$  are  
15 listed in Table 4 for the different particle types. Note that the activated fraction in Eq. 15 is  
16 limited to values between 0 and 1.

17 Although data were available only for ice supersaturation ranges below 25% (bacteria, illite  
18 NX, Saharan and Asian dust) and above 20% (illite IMt1 and feldspar), respectively, due to  
19 the investigated temperature ranges, it is assumed that Eq. 15 is valid for the complete ice  
20 supersaturation range. However, according to the onset of deposition freezing in the experi-  
21 ments, lower limits of ice supersaturation and temperature as measured during the FRIDGE  
22 experiments were set in the model. That means, deposition freezing of illite IMt1 and feldspar  
23 start at the same conditions as illite NX. In the model simulations, the two types of illite were  
24 taken as upper and lower limits and referred as illite 1 and illite 2. The values are given in  
25 Table 4.

### 26 **2.3.2 Treatment of deposition freezing in the model**

27 Conditions for deposition freezing are: (1) inactivated particles have to be present as it is  
28 required for contact freezing, i.e. the same particles may affect deposition or contact freezing.  
29 Additionally, (2) there are two size conditions. First, the inactivated particles have to exceed

1 a critical germ size  $r^*$  in dependence of temperature and ice supersaturation which is accord-  
2 ing to Pruppacher and Klett (2010):

$$3 \quad r^* = \frac{2 M_W \sigma_{i,v}}{RT \rho_{ice} \ln S_{ice}} \quad (16)$$

4 where  $M_W$  the molecular weight of water,  $\sigma_{i,v}$  the surface tension,  $R$  the universal gas constant,  
5  $T$  the temperature,  $\rho_{ice}$  the density of ice, and  $S_{ice}$  the ice saturation ratio. However, this condi-  
6 tion is actually redundant as it excludes particles smaller than approximately 0.01  $\mu\text{m}$  and  
7 additionally size restrictions of the ice nucleating particles were assumed: dust particles larger  
8 than 0.1  $\mu\text{m}$  in diameter and bacteria between their typical size range of 0.3 and 2  $\mu\text{m}$  in di-  
9 ameter (Matthias-Maser and Jaenicke, 1995).

10 In each time step it is checked if temperature and ice supersaturation are above the limit val-  
11 ues  $T_{ini}$  and  $S_{ice,ini}$ , if yes it is checked which available inactivated particles exceed the size  
12 limits, and from these the activated fraction is calculated. It is assumed that only a part  $F_{INP}$  of  
13 the available particles is able to act as ice nucleating particles. Therefore, the total number of  
14 particles in Eq. 15 is reduced:

$$15 \quad N_{act} = F_{INP} N_{total} \exp(a_{dep} + b_{dep} S_{ice}) \quad (17)$$

16 The activated particles are moved to the ice particle spectrum and grow further by water va-  
17 por deposition and they may serve as germs for secondary ice formation, i.e. they may initiate  
18 freezing of supercooled drops by collision (Diehl et al., 2006).

19

### 20 **3 Model initiation and sensitivity studies**

21 During the present sensitivity studies, convective clouds were simulated. In those clouds liq-  
22 uid drops are transferred into higher regions in the atmosphere and supercooled. Once frozen  
23 the ice particles grow further by riming (i.e. collision with other supercooled droplets) and by  
24 the deposition of water vapour (i.e. at the expense of liquid drops, the Bergeron-Findeisen  
25 process, which is typically much faster than condensation). Due to the vertical velocity large  
26 precipitation-sized ice particles can form in convective clouds and fall out as graupels, hail-  
27 stones or, when they fall through the melting layer, as large raindrops. The present model  
28 simulations were initialized with a convective vertical profile where temperatures in higher

1 altitudes were low enough to assure ice formation. It has an average lapse rate of approxi-  
2 mately 0.6 K per 100 m without any maxima or minima (Langmann, 2004), see Figure 7.

3 The ascent of the air parcel is driven by a temperature difference between the air bubble and  
4 its environment. Depending on the temperature difference the updraft of the air parcel pro-  
5 ceeds at various speeds and reaches various heights with corresponding temperatures (Diehl et  
6 al., 2006). For the present simulations, the final temperatures were  $-24.5^{\circ}\text{C}$ ,  $-29^{\circ}\text{C}$ , and  $-40^{\circ}\text{C}$   
7 with corresponding maximum altitudes of 9 km, 9.5 km, and 11 km; the maximum vertical  
8 velocities during the ascent of the parcel were  $15\text{ m s}^{-1}$ ,  $16.5\text{ m s}^{-1}$ , and  $19\text{ m s}^{-1}$ .

9 Two different dry aerosol particle number size distributions were used to compare the effects  
10 on ice formation. One was an average continental distribution (OPAC database, Hess et al.,  
11 1998) which is a rather broad spectrum and characterized by a large number of small parti-  
12 cles, see black line in Figure 8; its parameters are  $N = 7000\text{ cm}^{-3}$ ,  $d = 42.4\text{ nm}$ , and  $\sigma = 2.24$   
13 (with  $N$  the particle number,  $d$  the diameter, and  $\sigma$  the standard deviation). The other one was  
14 a regional haze distribution (Reid et al. 1998) which is a rather narrow spectrum and charac-  
15 terized by larger particle sizes. The parameters are  $N = 6000\text{ cm}^{-3}$ ,  $d = 0.1\text{ }\mu\text{m}$ , and  $\sigma = 1.65$ ,  
16 see red line in Figure 8. Both rather simple mono-modal distributions were selected to avoid a  
17 mixture of effects due to ice physics and activation of aerosol particles. Thus, the effects of  
18 ice physics should be emphasized. The soluble fraction  $\epsilon$  of the aerosol particles was set to 0.5  
19 which is a typical value of atmospheric particles (Busch et al., 2002).

20 Regarding the conditions on the sizes of the ice nucleating particles as given in Section 2  
21 (mineral dust particles larger than  $0.1\text{ }\mu\text{m}$ , bacteria larger than  $0.3\text{ }\mu\text{m}$ , pollen larger than  $2$   
22  $\mu\text{m}$ ), one can conclude from Figure 8 that – in particular in the average continental case – the  
23 majority of the continental particles is too small to affect ice formation. The dry particle spec-  
24 trum influences the drop spectrum which is important for immersion and contact modes. Fig-  
25 ure 9 shows the number concentrations as a function of diameter for two different altitudes  
26 and corresponding temperatures of zero and  $-29^{\circ}\text{C}$  for the two cases, average continental and  
27 regional haze particles. Because the liquid water content is the same during both model simu-  
28 lations, fewer but larger drops develop with the regional haze distribution and numerous but  
29 smaller drops with average continental distribution. On the other hand, as the continental  
30 spectrum is broader, collision and coalescence processes are more effective and, thus, larger  
31 drops evolve. Also given in Figure 9 are the number concentrations of the interstitial aerosol

1 particles. Note that during the updraft inactivated aerosol particles are available for ice for-  
2 mation in deposition contact modes.

3 With these initial conditions, a multitude of sensitivity studies was performed to demonstrate  
4 the impact of ice nuclei concentrations and types on ice formation in convective mixed-phase  
5 clouds. First, single freezing processes were studied while the following parameters were var-  
6 ied:

- 7 - dry aerosol particle number size distribution: average continental and regional haze,
- 8 - ice nucleating particle type – biological particles and mineral dust,
- 9 - temperature difference: 3K, 2K, and 1.5 K, leading to final temperatures of -40°C, -  
10 29°C, and -24.5°C, respectively,
- 11 - fraction of potential ice nucleating particles  $F_{INP}$  – variation between 0.001 and 10%.

12 Afterwards, coupled freezing processes were investigated to study the competition between  
13 the different freezing processes. These were undertaken only with those parameters which  
14 resulted in higher ice formation.

## 15 **4 Results and discussion**

### 16 **4.1 Ice water fractions and single freezing processes**

17 To evaluate the efficiency of the different freezing processes and ice nucleating particle  
18 types, as a central diagnostic parameter the ice water fraction  $IWF$  was selected which is cal-  
19 culated from the ice water content  $IWC$  and the liquid water content  $LWC$ :

$$20 \quad IWF = \frac{IWC}{LWC + IWC} \quad (19)$$

21 According to Korolev et al. (2003) an ice cloud is defined by  $IWF > 0.9$ , a liquid cloud by  
22  $IWF < 0.1$ , and mixed-phase clouds by  $0.1 \leq IWF \leq 0.9$ . Note that in an air parcel model, the  
23  $IWF$  is only influenced by in-situ ice formation processes and not by sedimentation of ice  
24 into or out of the considered parcel. In the following Tables 5 to 7, the ice water fractions as  
25 results from the sensitivity studies are listed for immersion, contact, and deposition freezing.

26 **Ice and mixed-phase clouds are marked by bold face type.**

27 A first sight on Table 5 to 7 shows that mixed-phase or ice clouds mainly evolved from the  
28 continental particle distribution with bacteria, feldspar, or illite INP and with larger  $\Delta T$ , i.e.

1 final temperatures reaching below  $-25^{\circ}\text{C}$ . The different points are discussed in detail in the  
2 following.

### 3 Particle number size distributions:

4 With the regional haze particles (larger particles but smaller numbers), less ice was formed in  
5 most cases than with the average continental particles (smaller particles but larger number).  
6 Less but mostly large drops develop with the regional haze distribution and many but smaller  
7 drops with average continental distribution (see Figure 9). On the other hand, a few still larg-  
8 er drops evolved from the tail of the continental spectrum. The presence of large drops favors  
9 immersion and contact freezing (immersion mode: the drops contain more insoluble material;  
10 contact mode: the collision kernel between large drops and particles is enhanced), see Tables  
11 5 and 6. This was suggested already by the findings of Diehl et al. (2006) and confirmed by,  
12 e.g., Lance et al. (2011) who concluded from their observations that the drop size distribution  
13 modulates ice processes in mixed-phase clouds.

14 In the deposition mode (Table 7), no drops but only the interstitial particles are relevant.  
15 More inactivated particles are present in the regional haze case than in the continental case  
16 (see Figure 9) so that because of the higher competition between the many particles, less ice  
17 particles develop by the deposition of water vapor.

### 18 Freezing modes:

19 Immersion freezing affects the most mixed-phase clouds with ice water fractions of more than  
20 0.5 and even ice clouds with all kinds of investigated INP (Table 5). Pollen are, of course,  
21 disadvantaged because of their large sizes, kaolinite particles are the less efficient mineral  
22 dust particles while bacteria and feldspar are the most efficient INP. With bacteria, freezing  
23 occurred already with potential fractions of INP  $F_{INP}$  as low as 0.001%. With mineral dust,  
24 potential fractions of INP  $F_{INP}$  of 0.01% (feldspar, illite) or 0.1% (kaolinite) were necessary.  
25 Ice formation was sensitive to the type of mineral dust as well as to the potential fraction of  
26 INP. For instance, under the same conditions an ice cloud could form with feldspar but a  
27 mixed-phase cloud with only a small ice water fraction with kaolinite.

28 In the contact mode, there is very little ice formation (Table 6). Only liquid clouds formed  
29 with the regional haze particle distribution but in case of the average continental distribution  
30 at least some mixed-phase clouds formed with ice water fractions between 0.1 and 0.3. High  
31 potential fractions of INP  $F_{INP}$  of 10% were required. Bacteria did not affect ice formation,

1 this probably results from the restricted particle sizes. The type of mineral dust decides  
2 whether mixed-phase clouds are formed (feldspar, montmorillonite) or not (illite, kaolinite).

3 Effective deposition INP are bacteria, feldspar, and illite while the mixed particle samples  
4 Saharan and Asian dust form liquid clouds only. Mixed-phase clouds formed with high poten-  
5 tial INP fractions  $F_{INP}$  between 1 and 10%. The results for the different mineral dust types are  
6 rather similar but mixed-phase clouds were formed with pure minerals only but not with mix-  
7 tures (Saharan and Asian dust) which are dominated by quartz (see Section 2.3.1).

8 Ervens et al. (2011) who investigated the impact of immersion and deposition freezing modes  
9 on ice formation in mixed-phase clouds stated that immersion freezing formed less ice than  
10 deposition freezing because of lower onset temperatures in the immersion mode. However,  
11 this could not be confirmed in the present study. Newer laboratory measurements which were  
12 used as basis of the parameterizations showed in contrary higher initial temperatures in the  
13 immersion mode than in the deposition mode (see Tables 1 and 4 with references therein) and,  
14 thus, immersion freezing produced more ice than deposition freezing. This agrees, on the oth-  
15 er hand, with the findings of de Boer et al. (2011) and Lance et al. (2011) that liquid-  
16 dependent ice nucleation modes are dominant.

#### 17 Temperature difference $\Delta T$ and final temperature

18 In all freezing modes, cases with the lowest temperature difference ( $\Delta T = 1.5K$ ) and the cor-  
19 responding highest final temperature of  $-24.5^{\circ}C$  showed hardly ice formation. Exceptions are  
20 only cases with bacteria and feldspar in the immersion mode (Table 5). Contact ice formation  
21 did not occur with lower  $\Delta T$  as the temperatures reached during the ascent of the air parcel  
22 were not low enough to give the more effective smaller particles the chance to act (Table 6).  
23 In the deposition mode, obviously temperatures below  $-25^{\circ}C$  are required to develop mixed-  
24 phase clouds (Table 7) as at higher temperatures the ice supersaturation is still too low. On the  
25 other hand, the largest  $\Delta T$  of 3K also hindered ice formation in contact and deposition modes  
26 which might be affected by the presence of less interstitial particles.

#### 27 **4.2 Comparison to measured INP numbers**

28 In this section a discussion is included how realistic the assumed concentrations of INP are  
29 which lead to partial or complete cloud glaciation.

#### 30 Immersion freezing

1 Regarding the cases of immersion freezing listed in Table 5, one has to look at the composi-  
2 tion of cloud residuals which was investigated in several field campaigns. E.g., Kamphus et  
3 al. (2010) measured 8% minerals in cloud droplet residuals; Hiranuma et al. (2013) found 3%  
4 mineral dust particles in cloud droplet residuals for all particle sizes and particle-size resolved  
5 measurements indicated enhanced fractions for larger particles up to 17%. In the model simu-  
6 lations mixed-phase clouds were formed already with potential INP fractions between 0.01  
7 and 1%. Thus, the dominant role of immersion freezing with mineral dust seems to be vali-  
8 dated. It is confirmed by the fact that during INUIT field campaigns (Worringen et al., 2014;  
9 Schmidt et al., 2015) fractions of mineral dust up to 40% in ice residuals were observed.  
10 Similar numbers between 30 and 40% are reported by Kamphus et al. (2010) in ice particle  
11 residuals.

12 Bacteria need to be present as potential INP only as low as 0.001 to 0.01% to affect mixed-  
13 phase clouds, pollen with 1% potential INP. For pollen these numbers do probably not repre-  
14 sent realistic cases; however, the pollen cases were anyway rather artificial as such large par-  
15 ticles ( $> 10 \mu\text{m}$ ) were actually not part of the aerosol particle spectrum. Bacteria concentra-  
16 tions in cloud water are given as average values of, e.g.,  $1.5 \times 10^9 \text{ m}^{-3}$  (Sattler et al., 2001),  $2$   
17  $\times 10^{10} \text{ m}^{-3}$  (Bauer et al. 2002), and  $7 \times 10^4 \text{ m}^{-3}$  (Amato et al., 2005; see also the review paper  
18 of Delort et al., 2010). Unfortunately, field measurements which give the number fractions of  
19 bacteria or primary biological aerosol particles (PBAP) related to the total concentrations of  
20 in-droplet particles are not available so far. An estimation was undertaken based on the results  
21 of Bauer et al. (2002). From field measurements in a continental background site they deter-  
22 mined the numbers of bacterial and fungi cells in cloud water and calculated the correspond-  
23 ing amount which would contribute to the amount of organic carbon (OC). For bacteria this  
24 contribution was estimated as 0.01%. Analyses of cloud droplet residuals show, e.g., fractions  
25 of 3% (Twohy and Anderson, 2008) and 9% (Hiranuma et al., 2013) organic carbon. Based  
26 on these numbers one could assume that 0.0003 to 0.0009% of the material contained in cloud  
27 drops consists of bacteria. This comes near the fraction of potential ice nucleating particles  
28  $F_{INP}$  of 0.001% which affects mixed-phase clouds via immersion freezing (see Table 5). Joly  
29 et al. (2014) investigated the ice nucleation efficiency of cloud water samples and distin-  
30 guished total and biological INP. They estimated that – assuming that all biological INP were  
31 bacteria – in the temperature range between  $-8$  and  $-12^\circ\text{C}$  0.6 to 3.1% of the bacterial cells  
32 present in the cloud water samples could have acted as INP. Taking into account these values,



1 a fraction  $F_{INP}$  of 0.001% is probably still an overestimation of realistic bacterial ice nucle-  
2 ating particles in cloud drops.

3 Thus, one may conclude that the atmospheric immersion freezing with mineral dust particles  
4 will play a dominant role while ice formation via immersion freezing of bacteria might take  
5 place in some extreme cases only.

### 6 Contact and deposition freezing

7 According to the cases listed in Tables 6 and 7, the formation of mixed-phase clouds via con-  
8 tact or deposition freezing requires that 10% of the background aerosol consists of potential  
9 ice nucleating particles. Measurements during an INUIT field campaign on the high alpine  
10 site Jungfrauoch (Schmidt et al., 2015) indicate a fraction of 1% minerals of the background  
11 aerosol; however, this value might be enhanced for continental situations in lower altitudes. A  
12 closer look was taken on the total number concentrations of ice nucleating particles acting as  
13 contact and deposition freezing particles during the model simulations. As shown in Figure 9  
14 for two different altitudes and corresponding temperatures, interstitial aerosol particles were  
15 always present during the ascent of the air parcel. Among these, only particles larger than 0.1  
16  $\mu\text{m}$  in diameter were allowed to act as ice nucleating particles. The total number concentra-  
17 tions of those during the model simulations reach values up to  $4 \times 10^5 \text{ m}^{-3}$  for the average  
18 continental and up to  $2 \times 10^6 \text{ m}^{-3}$  for the regional haze distribution. Thus, fractions of poten-  
19 tial ice nucleating particles  $F_{INP}$  of 10% resulted in particle concentrations available for depo-  
20 sition and contact nucleation of at the most  $4 \times 10^4 \text{ m}^{-3}$  and  $2 \times 10^5 \text{ m}^{-3}$ , respectively.

21 In Saharan dust events, Bangert et al. (2012) measured particle number concentrations up to  $5$   
22  $\times 10^7 \text{ m}^{-3}$ . In Hande et al. (2014), simulated mineral dust number concentrations are  $4 \times 10^5$   
23  $\text{m}^{-3}$  on average up to extreme values of  $5.8 \times 10^6 \text{ m}^{-3}$ . Thus, the numbers of potential INP used  
24 in the model simulations do not exceed realistic particle concentrations of mineral dust.

25 Near-surface concentrations of bacteria range between  $1 \times 10^3 \text{ m}^{-3}$  and  $5 \times 10^5 \text{ m}^{-3}$  depending  
26 on ecotypes (Burrows et al., 2009) but these values are certainly not reached in upper cloud  
27 regions. On the other hand, DeLeon-Rodriguez et al. (2013) reported from field measure-  
28 ments in low- and high-altitude air masses that bacterial cells represented nearly 20% of the  
29 total particles in the diameter range between 0.25 and 1  $\mu\text{m}$ . This is approximately the size  
30 range of potential ice nucleating particles in the present model simulations. For even larger  
31 particles in the coarse mode high fractions of PBAP (primary biological particles) are also

1 reported by Manninen et al. (2014). However, the numbers of potential INP used in the model  
2 simulations probably overestimate real bacteria concentrations.

3 Considering these factors one may conclude that the conditions for atmospheric deposition  
4 and contact freezing could be sufficient in some cases to form mixed-phase clouds from pri-  
5 mary ice formation by mineral dust particles. In some extreme cases, the formation of mixed-  
6 phase clouds might be possible via deposition nucleation on bacteria. On the other hand, the  
7 initial particle spectra used for the present model simulations contain little amounts of parti-  
8 cles larger than 1  $\mu\text{m}$  (see Figure 8) which would be able to act as contact ice nucleating par-  
9 ticles much more efficiently (see Section 2.2.1). Thus, in cases where larger INP are present  
10 in or around atmospheric clouds contact freezing might be significantly enhanced.

### 11 **4.3 Ice particle spectra, single and coupled freezing processes**

12 In this section only simulations with the most efficient ice formation are treated, i.e. using the  
13 average continental particle distribution and a medium  $\Delta T = 2\text{K}$  leading to final temperatures  
14 of  $-29^\circ\text{C}$ . The potential fractions  $F_{INP}$  were set to 10% for feldspar, illite, montmorillonite, and  
15 kaolinite. Bacteria and pollen were not considered here as those high values of  $F_{INP}$  are not  
16 realistic (see discussion in Section 4.2). Figure 10 shows the ice particle number size distribu-  
17 tions at different altitudes and corresponding temperatures for single deposition, contact, and  
18 immersion freezing processes which are discussed in the next paragraphs.

19 The ice particle spectra affected by immersion freezing are rather narrow starting with a parti-  
20 cle diameter of 1  $\mu\text{m}$  due to the smallest drop size. However, larger drops around 30  $\mu\text{m}$  in  
21 diameter were frozen first as their content of insoluble particles is higher. With decreasing  
22 temperature, also smaller drops can freeze while ice particles grow by the deposition of water  
23 vapour and by riming, i.e. the ice particle spectra broaden in both directions. Finally, they are  
24 still smaller than the ones formed by deposition freezing. The differences between the dust  
25 types are much more evident than in the other modes and the final ice particle numbers are  
26 higher, in particular for feldspar.

27 In the contact mode, the simulated ice particle spectrum is even somewhat narrower. At tem-  
28 peratures around  $-20^\circ\text{C}$ , the maximum of the drop number concentration lies at 30  $\mu\text{m}$ . This  
29 indicates that large supercooled drops froze by collisions with larger particles as in this tem-  
30 perature range smaller particles are hardly efficient as contact INP. Lowering the temperature  
31 down to  $-29^\circ\text{C}$  extends the ice particle spectra towards larger sizes, i.e. the ice particles grow

1 by the deposition of water vapor and by riming but still only little amounts of small drops are  
2 freezing. Thus, contact freezing is strongly controlled by the collision kernel. The number  
3 concentrations of ice particles formed on feldspar and montmorillonite are rather similar at -  
4 30°C but strongly different at -21°C. This is due to the fact that smaller feldspar particles are  
5 active at higher temperatures as smaller montmorillonite particles (see Figure 4).

6 The ice particle spectra due to deposition freezing start with small sizes and develop towards  
7 larger sizes during the ascent of the cloud. This indicates that firstly, as primary ice formation,  
8 small ice particles are formed due to the sizes of the involved particles (0.1  $\mu\text{m}$  at least **up to 3**  
9  **$\mu\text{m}$  maximum, see the initial particle spectra in Figure 8). Afterwards, these pristine ice parti-**  
10 **cles serve as nuclei for secondary ice formation, i.e. by collisions with supercooled liquid**  
11 **drops. This process produces ice particles of larger sizes, see maximum around 40  $\mu\text{m}$  in Fig-**  
12 **ure 10. Furthermore, all ice particles grow by the deposition of water vapor and by riming**  
13 **leading to ice particles larger than 100  $\mu\text{m}$ . Thus, a broad spectrum of ice particles evolved**  
14 **from deposition freezing. The number concentrations vary by one order of magnitude from**  
15 **illite2 to feldspar. The oscillations of the spectra on the left hand side are an artefact effect of**  
16 **the size classes of the particles acting as INP starting with 0.1  $\mu\text{m}$  diameter; they vanish if the**  
17 **lower size of the INP is limited by the critical radius only (Eq. 16).**

18 Coupled cases were simulated to investigate the competition between contact and immersion  
19 modes as both freeze supercooled drops, and the competition between contact and deposition  
20 freezing as both interact with inactivated particles. The following combinations were studied:  
21 1) feldspar and 2) illite1 as for these particle types parameterizations for all freezing modes  
22 are available based on INUIT measurements. Additionally, some mixed cases were investi-  
23 gated, i.e. one INP type for one freezing mode, another INP type for the other freezing mode.  
24 These were for coupled contact and immersion freezing 3) montmorillonite + kaolinite, 4)  
25 feldspar + kaolinite. **These cases were selected to combine weaker immersion freezing INP**  
26 **(kaolinite) with stronger contact freezing INP (feldspar and montmorillonite), see Figure 12a.**  
27 For coupled deposition and contact freezing the mixed cases were 3) illite2 + montmorillo-  
28 nite, 4) Saharan dust + feldspar. As can be noted from Tables 5 to 7, the ice water fractions  
29 evolved from feldspar INP are similar for deposition and contact freezing but significantly  
30 higher for immersion freezing. The effects from illite1 INP differ between the freezing modes  
31 (immersion highest, contact lowest). The combination illite2 + montmorillonite in deposition  
32 and contact modes was selected because the resulted ice water fractions were similar which is

1 also the case for the combination montmorillonite + kaolinite in contact and immersion  
2 modes.

3 In contrast, the fourth combinations Saharan dust + feldspar (deposition and contact) and feldspar + kaolinite (contact and immersion) were chosen as the ice water fractions evolved by  
4 contact freezing were higher. Figure 11 shows the ice particle number size distributions at  
5 different altitudes and corresponding temperatures for coupled deposition and contact and  
6 coupled contact and immersion freezing processes.  
7

8 One can note from Figure 11 that deposition and contact freezing are hardly in competition.  
9 The narrow ice particle spectrum due to contact freezing alone is enhanced towards smaller  
10 sizes due to deposition freezing. Small ice particles are formed by deposition freezing due to  
11 the sizes of the involved particles (0.1 to 3  $\mu\text{m}$ ), large ice particles are formed by contact  
12 freezing due to the sizes of involved liquid drops (30 to 50  $\mu\text{m}$ , see Figure 10). When first ice  
13 formation is observed via contact and deposition freezing at temperatures around  $-20^\circ\text{C}$ , in the  
14 contact mode only dust particles larger than 0.4  $\mu\text{m}$  (feldspar), 0.8  $\mu\text{m}$  (illite), and 1  $\mu\text{m}$  (kaolinite) are active (see Figure 4), thus, all smaller particles and fractions of the larger particles  
15 are available for deposition freezing. Even at lower temperatures, still small particles remain  
16 for deposition freezing as these are less efficient to collide with drops. The common ice particle  
17 spectra of coupled deposition and contact freezing are as broad as the ones from single  
18 deposition freezing but the number concentrations in the larger size range are enhanced in  
19 cases with efficient contact freezing, i.e. with feldspar and montmorillonite INP.  
20

21 Regarding coupled contact and immersion freezing, here the latter is the dominant process.  
22 Using the same particle types there is no effect visible from contact freezing (feldspar, illite1).  
23 However, in cases where the contact INP are more efficient than the immersion INP (feldspar  
24 or montmorillonite in contrast to kaolinite) the number concentrations are slightly enhanced in  
25 the size range larger than 90  $\mu\text{m}$  (see Figure 10 and 11).

26 The total numbers of liquid drops and ice particles in  $\text{cm}^{-3}$  as a function of temperature are  
27 given in Figure 12. Results from single freezing processes are shown in Figure 12a. In the  
28 upper part of the figure, liquid drop numbers are shown as solid lines. The black line represent  
29 liquid drop numbers in the order of  $8 \times 10^2 \text{ cm}^{-3}$  which applies for all cases except immersion  
30 freezing with feldspar. Below, in all remaining plot lines ice particle numbers are given in  
31 Figure 12a for the three single freezing processes as colored lines (the colors describe the  
32 types of INP) with different line styles: solid lines for immersion freezing (feldspar, illite1,

1 and kaolinite), dotted lines for contact freezing (feldspar, montmorillonite, and illite), and  
2 broken lines for deposition freezing (feldspar, illite1, illite2, and Saharan dust).

3 From the ice particle numbers at the lowest temperature of  $-29^{\circ}\text{C}$ , one can clearly distinguish  
4 between cloud types: ice particle numbers around  $1\text{ cm}^{-3}$  represent an ice cloud (immersion  
5 freezing with feldspar), ice particle numbers around  $1 \times 10^{-8}\text{ cm}^{-3}$  represent a liquid cloud (i.e.  
6 the ice water fraction is smaller than 0.1; contact freezing with illite), and ice particle numbers  
7 between  $1 \times 10^{-4}\text{ cm}^{-3}$  and  $1 \times 10^{-2}\text{ cm}^{-3}$  represent mixed-phase clouds (all other cases). In the  
8 only case of ice cloud formation (immersion with feldspar) the liquid drop number is affected  
9 as well: it is reduced to  $5 \times 10^1\text{ cm}^{-3}$  (i.e. the ice water fraction is larger than 0.9), see red solid  
10 line in the upper part of the figure.

11 It can be seen that the development of ice particle numbers with decreasing temperature via  
12 immersion freezing (colored solid lines) is similar to the development of the numbers of ac-  
13 tive sites immersed in the drops with decreasing temperature (shown in Figure 1). In contrast,  
14 contact freezing (dotted lines) is not ruled by the temperature alone but also by the collision  
15 efficiencies between potential INP and supercooled drops (see Section 2.2) while deposition  
16 freezing (broken lines) is ruled by the ice supersaturation (see Section 2.3) which, of course,  
17 increases with decreasing temperature.

18 Comparing the liquid drop numbers to the ice particle numbers one notes that the differences  
19 are approximately between 4 and 6 orders of magnitude in cases of mixed-phase clouds and  
20 still more than one order of magnitude in case of the ice cloud. This indicates that the glacia-  
21 tion of the clouds proceeds mainly by the growth of ice particles at the expense of liquid  
22 drops (Bergeron-Findeisen process or riming). Here, some effects are presumably overesti-  
23 mated as from the air parcel no cloud particle sedimentation is possible.

24 Figure 12b shows results for the coupled freezing processes. Again liquid drop numbers are  
25 given as solid lines in the upper part of the figure. The black line represents liquid drop num-  
26 bers which applies for all cases except immersion + contact freezing with feldspar. Lower plot  
27 lines in Figure 12b show ice particle numbers for the two coupled freezing processes as col-  
28 ored lines (the colors describe the types of INP) with different line styles: solid lines for im-  
29 mersion + contact freezing (feldspar, illite1, kaolinite + feldspar, kaolinite + montmorillonite),  
30 and broken lines for deposition + contact freezing (feldspar, illite1, illite2 + montmorillonite,  
31 Saharan dust + feldspar).

1 Ice clouds and mixed-phase clouds differ in the ice particle numbers at the lowest temperature  
2 of  $-29^{\circ}\text{C}$ : ice particle numbers around  $1\text{ cm}^{-3}$  represent an ice cloud (immersion + contact  
3 freezing with feldspar), and ice particle numbers between  $5 \times 10^{-4}\text{ cm}^{-3}$  and  $1 \times 10^{-2}\text{ cm}^{-3}$  rep-  
4 resent mixed-phase clouds (all other cases). In the only case of ice cloud formation the liquid  
5 drop number is affected as well, see red solid line in the upper part of the figure.

6 It is obvious that in those cases where one process is inferior the ice particle numbers are  
7 completely determined by the dominant process. This is the case in coupled immersion and  
8 contact freezing with feldspar and illite1 but not with kaolinite + feldspar and kaolinite +  
9 montmorillonite where the effects of contact freezing are visible at lower temperatures ( $< -$   
10  $20^{\circ}\text{C}$ ). Coupled deposition and contact freezing shows the same results as for deposition  
11 freezing alone in case of illite. In the other cases, feldspar, illite2 + montmorillonite, and Sa-  
12 haran dust + feldspar, there are at least some small enhancements visible at temperatures low-  
13 er than  $-20^{\circ}\text{C}$  when contact freezing becomes more efficient.

#### 14 **4.4 Comparison of presently simulated ice particle numbers to previous litera-** 15 **ture data**

16 In this section results from the model simulations are compared to data from previous model  
17 simulations and atmospheric measurements. As comparison parameter the number of ice parti-  
18 cles formed at the end of the air parcel ascent was used as shown in Figure 12. These simu-  
19 lations were performed with the continental particle distribution,  $dT=2\text{K}$ , and  $F_{INP}=10\%$ . I.e.  
20 for  $F_{INP}$  values lower than 10% less ice particle numbers are formed.

21 Eidhammer et al. (2009) used an air parcel model to perform intercomparisons of three heter-  
22 ogeneous ice nucleation parameterizations which linked aerosol types and numbers to ice parti-  
23 cle number concentrations. One of these was the previous immersion freezing parameteriza-  
24 tion of Diehl and Wurzler (2004) which has been replaced by a new one in this study. This de-  
25 scription was related to the volume of liquid similar as the well-known Bigg parameterization  
26 (Bigg, 1953). The criticism was that some constraints are required that limit the number of  
27 potential ice nucleating particles. The present parameterization is related to the insoluble par-  
28 ticle mass contained in the drops and the constraint is given by the factor  $F_{INP}$  in Eq. 8. Ice  
29 particle numbers found by Eidhammer et al. (2009) with the Diehl and Wurzler (2004) de-  
30 scription were in the range of  $5 \times 10^{-2}$  to  $5 \times 10^{-1}\text{ cm}^{-3}$  while results from the current simula-  
31 tions are in the order of  $1 \times 10^{-3}\text{ cm}^{-3}$  for mixed-phase clouds (see Figure 12, immersion freez-  
32 ing).

1 In Ervens and Feingold (2012) the effects of five ice nucleation schemes for immersion freez-  
2 ing with particles showing surface characteristics like kaolinite particles on the properties of  
3 mixed-phase clouds were compared in air parcel studies. The present parameterization of im-  
4 mersion freezing is similar to the so-called deterministic scheme of Ervens and Feingold  
5 (2012) which includes a cumulative activation spectrum as a function of temperature. Using  
6 this scheme in their parcel model with polydisperse INP, they found ice particle numbers  
7 from  $2 \times 10^{-4}$  to  $1 \times 10^{-3} \text{ cm}^{-3}$ . In spite of differences between the present and previous air  
8 parcel studies (e.g., height of parcel elevation) the present results of immersion freezing with  
9 kaolinite particles, i.e.  $3.5 \times 10^{-4} \text{ cm}^{-3}$  (see Figure 12a) agree very well.

10 Fan et al. (2010) simulated two deep convective clouds from field campaigns, one cloud  
11 formed under clean conditions, the other under polluted conditions with biomass burning.  
12 They used a three-dimensional cloud-resolving model with different immer-  
13 sion/condensation/deposition and homogeneous ice nucleation parameterizations. The aver-  
14 aged cloud properties for their clean case showed ice particle numbers between  $1.3 \times 10^{-2}$  and  
15  $1.5 \times 10^{-2} \text{ cm}^{-3}$  in dependence of the ice nucleation schemes. For the comparison coupled im-  
16 mersion and deposition freezing was simulated with kaolinite acting in the immersion mode,  
17 Saharan dust in the deposition mode (not shown in Figure 12b). These two particle types  
18 might represent the behaviour of “mean” atmospheric INP. During the calculations  $dT$  was  
19 varied by 2K, 2.5K, and 3K. The final ice particle numbers were  $4.8 \times 10^{-4} \text{ cm}^{-3}$ ,  $2.2 \times 10^{-2}$   
20  $\text{cm}^{-3}$ , and  $6.4 \times 10^{-1} \text{ cm}^{-3}$ , respectively, and, thus, best agreement with a deep convective cloud  
21 was obtained with  $dT=2.5\text{K}$ .

22 Kulkarni et al. (2012) used Arizona test dust (ATD) and kaolinite to experimentally investi-  
23 gate and model deposition freezing. However, their measured activated fractions of particles  
24 as a function of ice supersaturation were at least one to two orders of magnitude larger than  
25 the ones used as basis for the parameterizations in the present study. Therefore, not surpris-  
26 ingly, the modeled ice particle numbers found by Kulkarni et al. (2012) were between  $2 \times 10^{-3}$   
27 and  $1 \times 10^{-2} \text{ cm}^{-3}$  and up to two orders of magnitude larger than the ones found in the present  
28 simulations ( $1.4 \times 10^{-4} \text{ cm}^{-3}$  for Saharan dust,  $7.2 \times 10^{-4} \text{ cm}^{-3}$  for illite2,  $1.8 \times 10^{-3} \text{ cm}^{-3}$  for  
29 illite1, and  $4.5 \times 10^{-3} \text{ cm}^{-3}$  for feldspar, see Figure 12a).

30 Observations of atmospheric clouds reported ice particle number concentrations of  $1 \times 10^{-4}$  to  
31  $10 \text{ cm}^{-3}$  in convective clouds (Hobbs et al., 1980),  $1 \times 10^{-5}$  to  $5 \times 10^{-2} \text{ cm}^{-3}$  in mixed phase  
32 clouds (McFarquhar et al.; 2007; modeled by Fridlind et al., 2007);  $1 \times 10^{-1}$  to  $1 \text{ cm}^{-3}$  in ice

1 clouds (Krämer et al., 2009). Thus, the ice particle numbers formed during the present model  
2 simulations, i.e.  $1 \times 10^{-4}$  to  $1 \times 10^{-2} \text{ cm}^{-3}$  for mixed-phase clouds and  $9 \times 10^{-1} \text{ cm}^{-3}$  for ice  
3 clouds (see Figure 12) represent the status of real atmospheric clouds.

## 4 **5 Summary and conclusions**

5 In this paper improvements and modifications of the spectral-bin microphysics embedded in  
6 an adiabatic air parcel model with entrainment as described in Diehl et al. (2006) are present-  
7 ed. They include (1) a particle-type dependent parameterization of immersion freezing, (2) a  
8 particle-type and size-resolved parameterization of contact freezing, and (3) a particle-type  
9 dependent description of deposition freezing. Sensitivity studies with the modified version of  
10 the microphysics package demonstrated the impact of ice nuclei concentrations and types on  
11 ice formation in convective mixed-phase clouds. Single and coupled freezing processes were  
12 studied and the following parameters were varied: initial aerosol particle number size distribu-  
13 tions, final temperature, types of INP, and the fractions of potential INP.

14 The majority of mixed-phase and ice clouds were formed at temperatures below  $-25^\circ\text{C}$ , with  
15 the average continental particle number size distribution and with immersion freezing, even  
16 with smaller values of  $F_{INP}$  (fractions of potential INP).

17 First, larger drops freeze in the immersion mode because they contain more particulate mate-  
18 rial. During the updraft of the cloud and corresponding lower temperatures, the ice particle  
19 spectra develop towards smaller sizes. Contact freezing is limited by the collision kernel be-  
20 tween supercooled drops and potential ice nucleating particles. Large particles collide more  
21 often with large drops. Because they are more efficient contact INP than smaller particles this  
22 leads at first to the formation of larger ice particles. Smaller particles which represent in gen-  
23 eral the majority of the particle spectrum become effective at lower temperatures and, thus,  
24 contact freezing becomes relevant below  $-25^\circ\text{C}$ . During coupled immersion and contact freez-  
25 ing immersion freezing is the dominant process in cases with the same INP types (feldspar,  
26 illite). In mixed cases where contact INP are more efficient than immersion INP (feldspar or  
27 montmorillonite with kaolinite), contact freezing contributes similarly as immersion freezing  
28 to ice formation. In such cases sufficient liquid drops are available for contact freezing.

29 The importance of deposition freezing lies in secondary ice formation: at first, small pristine  
30 ice particles are formed due to the sizes of the involved particles which trigger the freezing of  
31 supercooled drops by collisions. Thus, a broader ice particle spectrum is generated than by



1 immersion and contact freezing. Regarding coupled contact and deposition freezing, there is  
2 hardly competition because both start moderate at higher temperatures so that inactivated par-  
3 ticles are present for both freezing modes.

4 The most effective ice nucleating particles are bacteria, feldspar, and illite; of minor im-  
5 portance are pollen, montmorillonite, and kaolinite. Ice formation by immersion freezing is  
6 very sensitive to the different mineral dust types leading to ice particle numbers varying by  
7 three orders of magnitude.

8 From the model results it can be concluded that the formation of mixed-phase and ice clouds  
9 in convective situations is promoted by (1) the immersion freezing mode, (2) broad drop size  
10 spectra containing small as well as large drops, (3) insoluble particles composed by bacteria,  
11 feldspar, and illite, and (4) temperatures below  $-25^{\circ}\text{C}$ .

12 The dominance of the immersion mode confirms the findings of de Boer et al. (2011) and  
13 Lance et al. (2011) about the importance of liquid-dependent ice nucleation modes. The role  
14 of contact freezing remained still unclear. In Diehl et al. (2006) it was estimated to be the  
15 most efficient process; however, this was because of assuming high freezing efficiencies for  
16 all particle sizes. Thus, the effects in the present model simulations with the modified contact  
17 freezing description should be closer to the real situation in the atmosphere. Contact freezing  
18 might be enhanced in atmospheric situations where particles larger than  $1\ \mu\text{m}$  are present in  
19 higher amounts. The dependence of ice formation on the properties of the liquid drop size  
20 spectrum was also observed by Lance et al. (2011). This should be investigated in more detail  
21 but was not the main focus of the present simulations which concentrated more on the impact  
22 of ice nucleating particle types.

23 Bacteria as primary biological particles were found to affect mixed-phase or even ice clouds;  
24 however, the critical factor is the availability in atmospheric environment and clouds. Togeth-  
25 er with estimations of their atmospheric occurrence the present model simulations indicate  
26 that they are probably not involved in significant ice formation. This has been suggested al-  
27 ready by Phillips et al. 2009, Diehl and Wurzler (2010), and Paukert and Hoose (2014). Thus,  
28 mineral dust particles seem to be the most important INP. The model results indicate that ice  
29 formation by immersion freezing is similarly sensitive to the mineral dust types as to the po-  
30 tential fractions of INP. Therefore, the composition of dust particles decides their impact on  
31 ice nucleation in clouds; essential components are feldspar and illite. In particular, the inves-  
32 tigation of typical atmospheric mixtures of mineral dust is relevant.

1 The microphysical package presented here was included into an air parcel model which has  
2 the advantage that all changes in the microphysical evolution of the cloud can be attributed to  
3 microphysical processes. On the other hand, some compromises are required concerning the  
4 cloud dynamics including some well-known weaknesses as precipitation sized cloud particles  
5 do not sediment but stay inside the parcel. As they are not removed from the parcel they could  
6 grow to unrealistic sizes. This can happen notably in the ice phase and lead to an overestima-  
7 tion of cloud glaciation. Some more limitations which should be considered are related to  
8 immersion and contact freezing. During the cloud model runs each drop contains insoluble  
9 material serving as immersion ice nucleus because the particles in the model are internally  
10 mixed. In a real cloud there might be drops which do not contain any potential ice nucleating  
11 material. Furthermore, the values of  $F_{INP}$  used in the model represent maximum estimations as  
12 not all dust particles are able to affect freezing at atmospheric temperatures. For contact freez-  
13 ing, the amount of interstitial inactivated particles is limited in an air parcel model while in a  
14 real cloud there might be more inactivated particles at the edges or beneath the cloud.

15 The further goal is to implement the new microphysical scheme into to a more complex state-  
16 of-the-art model system. For this purpose presently the 3D cloud model COSMO-SPECS  
17 (Grützun et al, 2008) is employed. This model contains the microphysical scheme as used in  
18 Diehl et al. (2006) within the adiabatic parcel model which is replaced by the microphysical  
19 scheme presented here. Such an improvement will allow more complex model simulations  
20 including the formation of precipitation which will enlighten the role of ice nucleating parti-  
21 cles in atmospheric clouds.

22

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30

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particle type	$a_{imm}$	$b_{imm}$	$T_{ini}$ °C	$T_{lim}$ °C
bacteria (1)	6.41344	2.33592	-2	-9.139
tree pollen (2;3)	7.16249	0.62053	-9	-38
grass pollen (2;3)	9.9731	0.030301	-12	-38
feldspar (4)	2.10379	1.038	-5	-25
illite (5)	-1.77473	0.89507	-10	-37
kaolinite (6)	-4.61608	0.8881	-13	-37

2 Table 1: Values of immersion freezing constants in Eq. 2. Based on data from (1) Wex et al.,  
3 2015; (2) Diehl et al., 2002; (3) v. Blohn et al., 2005; (4) Atkinson et al., 2013; (5) Hiranuma  
4 et al., 2015; (6) Murray et al., 2011.

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particle type and size	$a_{con}$	$b_{1,con}$	$b_{2,con}$	$b_{3,con}$
<b>bacteria</b>				
(1) $0.3 \mu\text{m} \leq d_{ap} < 0.5 \mu\text{m}$	-1.36581	-0.26367	-0.01511	-2.84911
(1) $0.5 \mu\text{m} \leq d_{ap} < 0.7 \mu\text{m}$	-0.55381	-0.10712	-0.00616	-1.16822
	$a_{con}$	$b_{con}$		
(2) $0.7 \mu\text{m} \leq d_{ap} < 2 \mu\text{m}$	-0.264	-0.742		

8 Table 2: Values of contact freezing constants in Eqs. 10 and 11. Based on data from (1)  
9 Hoffmann and Kiselev, 2014, who used Snomax<sup>®</sup> as a proxy for bacteria and (2) Levin and  
10 Yankofsky, 1983. The latter values were used in Diehl et al., 2006.

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particle type and size	$a_{con}$	$b_{con}$
<b>illite</b>		
(1) $0.1 \mu\text{m} < d_{ap} \leq 0.2 \mu\text{m}$	-0.0306	-0.9980
(1) $0.2 \mu\text{m} < d_{ap} \leq 0.4 \mu\text{m}$	-0.0359	-1.0838
(1) $0.4 \mu\text{m} < d_{ap} \leq 0.6 \mu\text{m}$	-0.0812	-2.2989
(1) $0.6 \mu\text{m} < d_{ap} \leq 0.8 \mu\text{m}$	-0.1210	-3.3452
(2) $0.8 \mu\text{m} < d_{ap}$	-0.1863	-1.6076
<b>montmorillonite</b>		
(3) $0.1 \mu\text{m} < d_{ap} \leq 1 \mu\text{m}$	-0.1014	-2.3899
(4) $1 \mu\text{m} < d_{ap}$	-0.1014	-0.3277
<b>kaolinite</b>		
(5) $0.1 < d_{ap} \leq 1 \mu\text{m}$	-0.1285	-3.6792
(6) $1 \mu\text{m} < d_{ap}$	-0.1007	-0.6935
<b>feldspar</b>		
(7) $0.1 < d_{ap} \leq 0.4 \mu\text{m}$	-0.1033	-2.0585
(7) $0.4 < d_{ap} < 0.8 \mu\text{m}$	-0.1484	-2.7154
(8) $0.8 \mu\text{m} < d_{ap}$	-0.34964	-0.13095

1 Table 3: Values of contact freezing constants in Eq. 10. Based on data from (1) Hoffmann et  
2 al., 2013b; (2) Diehl, 2012; (3) Extrapolation; (4) and (6) Pitter and Pruppacher, 1973, used in  
3 Diehl et al., 2006; (5) Hoffmann et al. (2013a); (7) Hoffmann and Kiselev, 2014; (8) Diehl  
4 and Mitra (2014).

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particle type	$a_{dep}$	$b_{dep}$	$T_{ini}$ °C	$S_{ice,ini}$ %
bacteria (1)	-12.65977	0.33382	-10 (1)	3 (1)
feldspar (2)	-14.58404	0.23576	-13 (1)	6 (1)
illite 1 (1)	-12.79648	0.15451	-13 (1)	6 (1)
illite 2 (2)	-15.11871	0.19669	-13 (1)	6 (1)
Saharan dust (3)	-13.39669	0.10058	-18 (3)	11 (3)
Asian dust (3)	-14.98495	0.09756	-15 (3)	8 (3)

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Table 4: Values of deposition freezing constants  $a_{dep}$  and  $b_{dep}$  in Eq. 15 and values of lower limits of temperature  $T_{ini}$  and ice supersaturation  $S_{ice,ini}$ . Based on data from (1) Danielczok and Bingemer, 2014; (2) Yakobi-Hancock et al., 2013; (3) Ardon-Dryer and Levin, 2012.

immersion freezing										
AP	regional haze					average continental				
$F_{INP}$ (%)	0.001	0.01	0.1	1	10	0.001	0.01	0.1	1	10
<b>dT = 3K    T<sub>fin</sub> = -40°C</b>										
bacteria	0.27	0.94	0.99	1.0	1.0	0.03	0.27	0.91	0.99	1.0
pollen	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02	0.08	0.15	0.17
feldspar	< 0.01	0.12	0.74	1.0	1.0	< 0.01	0.10	0.37	0.93	1.0
illite	< 0.01	0.01	0.14	1.0	1.0	< 0.01	0.19	0.43	0.70	1.0
kaolinite	< 0.01	< 0.01	< 0.01	0.64	0.81	< 0.01	< 0.01	0.12	0.34	0.60
<b>dT = 2K    T<sub>fin</sub> = -29°C</b>										
bacteria	0.26	0.86	0.99	1.0	1.0	0.13	0.43	0.86	0.99	1.0
pollen	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02	0.13	0.33
feldspar	< 0.01	0.02	0.21	0.71	1.0	0.08	0.34	0.56	0.80	0.93
illite	< 0.01	< 0.01	< 0.01	< 0.01	0.02	< 0.01	< 0.01	0.04	0.24	0.47
kaolinite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02	0.15
<b>dT = 1.5K    T<sub>fin</sub> = -24.5°C</b>										
bacteria	0.11	0.61	0.96	1.0	1.0	0.05	0.25	0.69	0.97	1.0
pollen	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.03
feldspar	< 0.01	< 0.01	< 0.01	0.03	0.24	< 0.01	< 0.01	0.03	0.35	0.59
illite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.01
kaolinite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01

2 Table 5: Ice water fractions from sensitivity studies with immersion freezing. Liquid clouds:  
3  $IWF < 0.1$ , mixed-phase clouds:  $0.1 \leq IWF \leq 0.9$ , ice clouds:  $IWF > 0.9$ .

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<b>contact freezing</b>						
AP	regional haze			average continental		
$F_{INP}$ (%)	0.1	1	10	0.1	1	<b>10</b>
<b>dT = 3K      T<sub>fin</sub> = -40°C</b>						
bacteria	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
feldspar	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.06
montmorillonite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.04
illite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
kaolinite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
<b>dT = 2K      T<sub>fin</sub> = -29°C</b>						
bacteria	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
<b>feldspar</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	<b>0.23</b>
<b>montmorillonite</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	<b>0.12</b>
illite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
kaolinite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
<b>dT = 1.5K      T<sub>fin</sub> = -24.5°C</b>						
bacteria	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
feldspar	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.01
montmorillonite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
illite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
kaolinite	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01

Table 6: Ice water fractions from sensitivity studies with contact freezing. Liquid clouds:  $IWF < 0.1$ , mixed-phase clouds:  $0.1 \leq IWF \leq 0.9$ , ice clouds:  $IWF > 0.9$ .



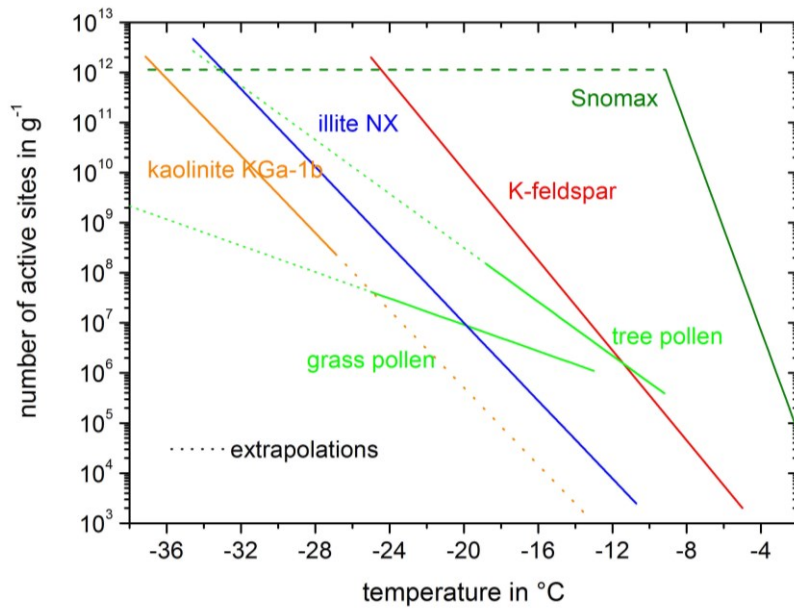
deposition freezing								
AP	regional haze				average continental			
$F_{INP}$ (%)	0.01	0.1	1	<b>10</b>	0.01	0.1	<b>1</b>	<b>10</b>
<b>dT = 3K    T<sub>fin</sub> = -40°C</b>								
<b>bacteria</b>	< 0.01	< 0.01	0.05	<b>0.52</b>	< 0.01	0.02	<b>0.14</b>	<b>0.42</b>
<b>feldspar</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02	<b>0.10</b>	<b>0.31</b>
<b>illite 1</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02	<b>0.13</b>
<b>illite 2</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.01	<b>0.10</b>
Saharan dust	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.01
Asian dust	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
<b>dT = 2K    T<sub>fin</sub> = -29°C</b>								
<b>bacteria</b>	< 0.01	< 0.01	< 0.01	0.06	< 0.01	0.04	<b>0.27</b>	<b>0.51</b>
<b>feldspar</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.07	<b>0.34</b>
<b>illite 1</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.03	<b>0.22</b>
<b>illite 2</b>	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.01	<b>0.11</b>
Saharan dust	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	0.02
Asian dust	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
<b>dT = 1.5K    T<sub>fin</sub> = -24.5°C</b>								
bacteria	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
feldspar	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
illite 1	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
illite 2	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Saharan dust	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Asian dust	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01

1 Table 7: Ice water fractions from sensitivity studies with deposition freezing. Liquid clouds:  
2  $IWF < 0.1$ , mixed-phase clouds:  $0.1 \leq IWF \leq 0.9$ , ice clouds:  $IWF > 0.9$ .

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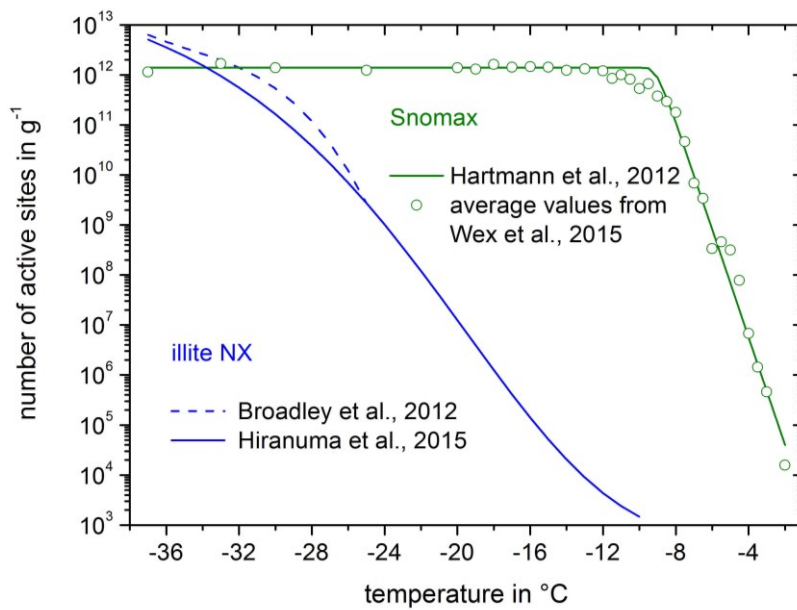
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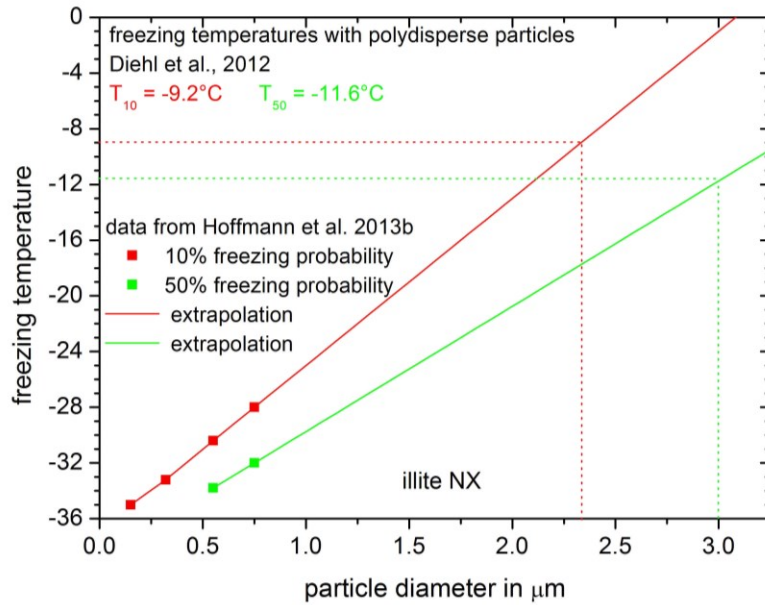
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Figure 1: Numbers of active sites per unit mass as a function of temperature calculated by Eq. 2 with constants given in Table 1 for various particle types in the immersion mode. Based on laboratory data of immersion freezing experiments (Diehl et al., 2002, v. Blohn et al., 2005, pollen; Murray et al., 2011, kaolinite KGa-1b; Atkinson et al., 2013, K-feldspar; Hiranuma et al., 2015, illite NX; Wex et al., 2015, Snomax<sup>®</sup>).



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Figure 2: Numbers of active sites per unit mass as a function of temperature. Parameterizations for illite NX and Snomax<sup>®</sup> based on the outcome of laboratory immersion freezing experiments.

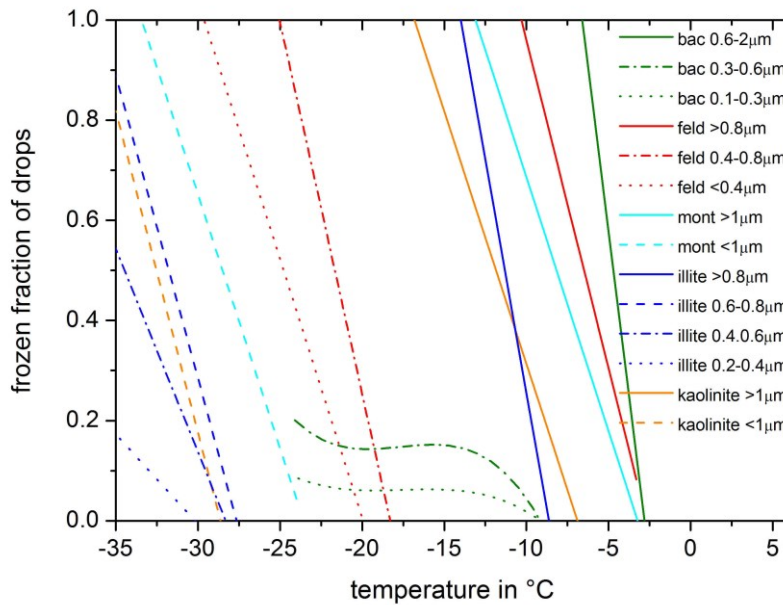


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2 Figure 3: Freezing temperatures as a function of particle diameter for illite NX in the contact  
 3 mode. Extrapolations to larger particle sizes.

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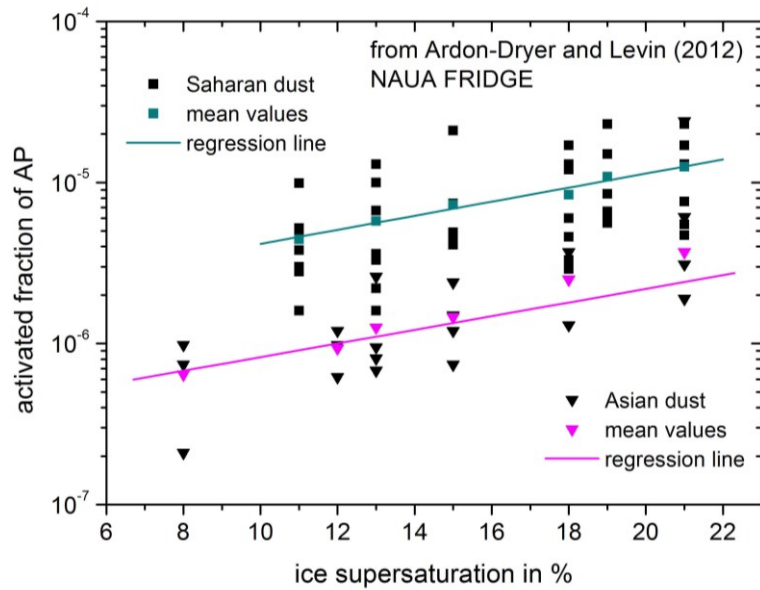
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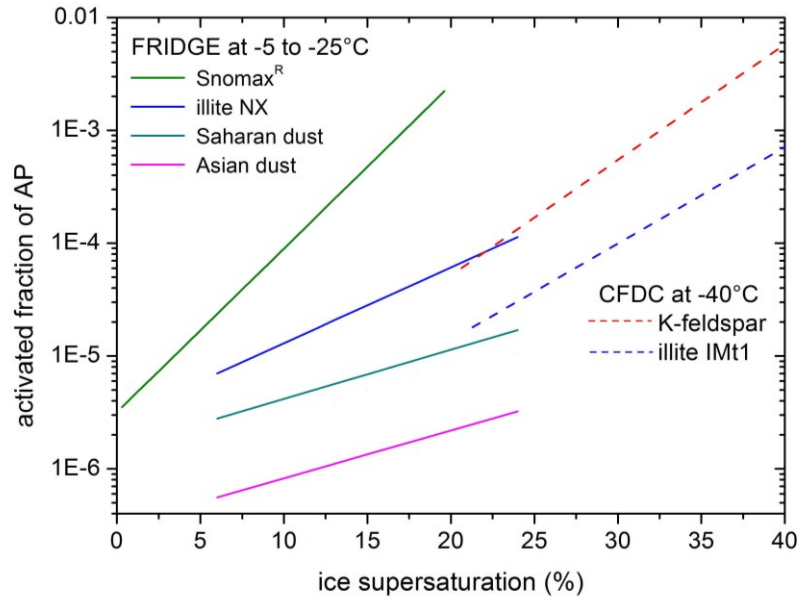
7 Figure 4: Frozen fraction of drops by contact freezing as a function of temperature for various  
 8 particle types and sizes, marked by different colors and line styles. Calculated by Eqs. 10 and  
 9 11, respectively, with constants given in Tables 2 and 3 for various particle types, based on  
 10 the outcome of laboratory experiments (Pitter and Pruppacher, 1973; Levin and Yankofsky,  
 11 1983; Diehl, 2012; Hoffmann et al., 2013a,b; Hoffmann and Kiselev, 2014; Diehl and Mitra,  
 12 2014).

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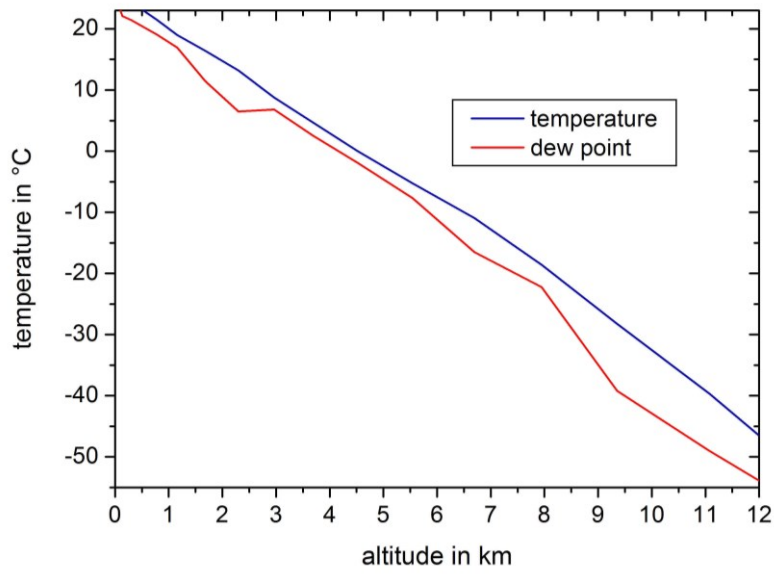
Figure 5: Activated fraction of particles **in the deposition freezing mode** as a function of ice supersaturation for Saharan and Asian dust. Data from Ardon-Dryer and Levin (2012).



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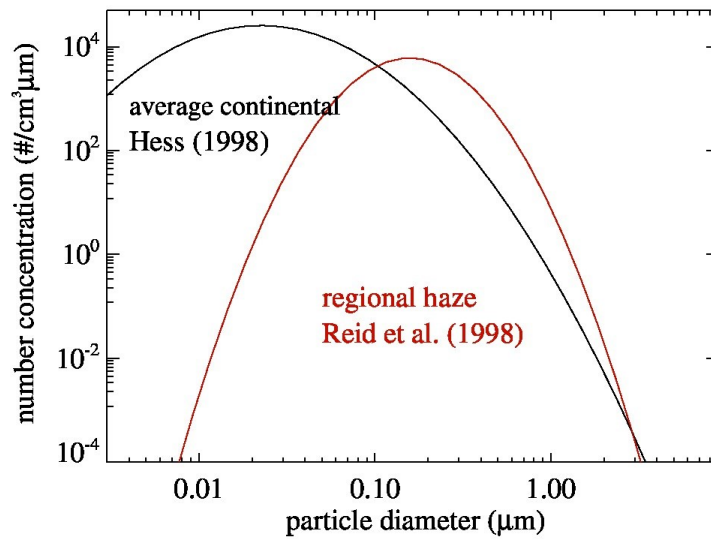
Figure 6: Activated fraction of particles **in the deposition freezing mode** as a function of ice supersaturation for various particle types. Regression lines based on data from Ardon-Dryer and Levin (2012; Saharan and Asian dust), Yakobi-Hancock et al. (2013; K-feldspar and illite IMt1), Danielczok and Bingemer (2014, Snomax<sup>®</sup> and illite NX). **Described by Eq. 15 with constants in Table 4.**

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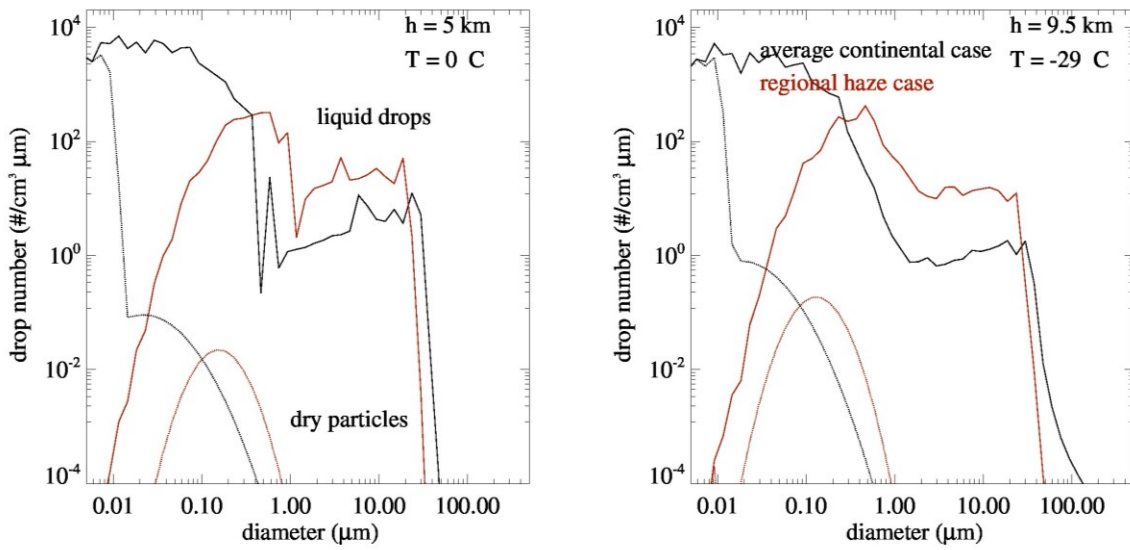
Figure 7: Development of temperature and dew point with altitude during simulations with the air parcel model (Langmann, 2004).



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Figure 8: Initial dry aerosol particle number size distributions: Number concentrations per cm<sup>3</sup> and μm as a function of particle diameter.

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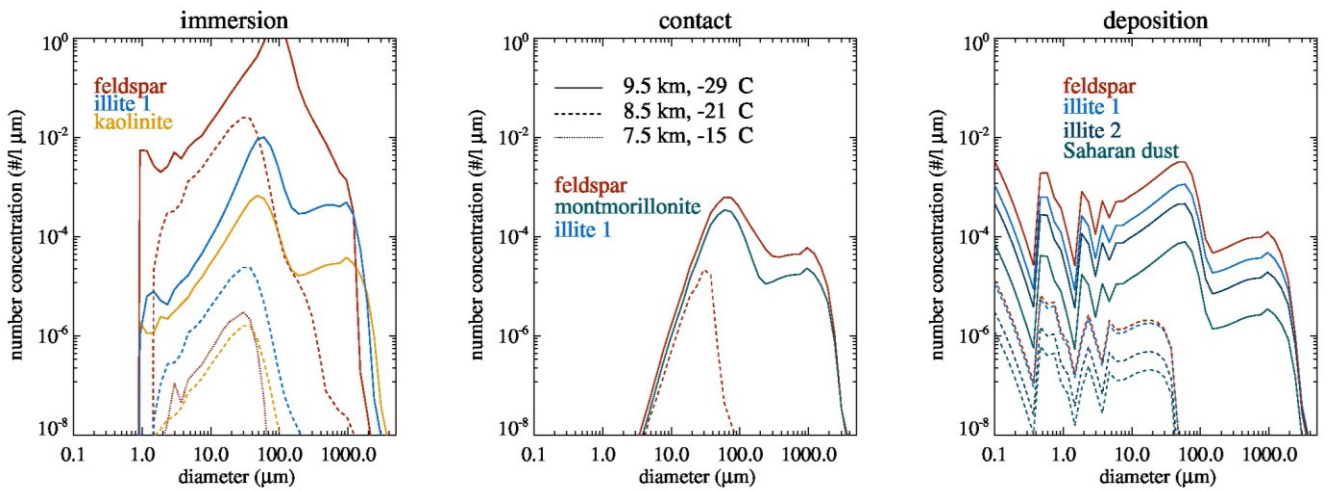
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3 Figure 9: Results from model runs without freezing. Development of liquid drop (solid lines)  
4 and interstitial particle numbers (dotted lines) with altitude for two initial aerosol particle  
5 number size distributions, average continental (black lines), regional haze (red lines), with  $\Delta T$   
6 = 2K. Number concentrations per  $\text{cm}^3$  and  $\mu\text{m}$  as a function of particle diameter.

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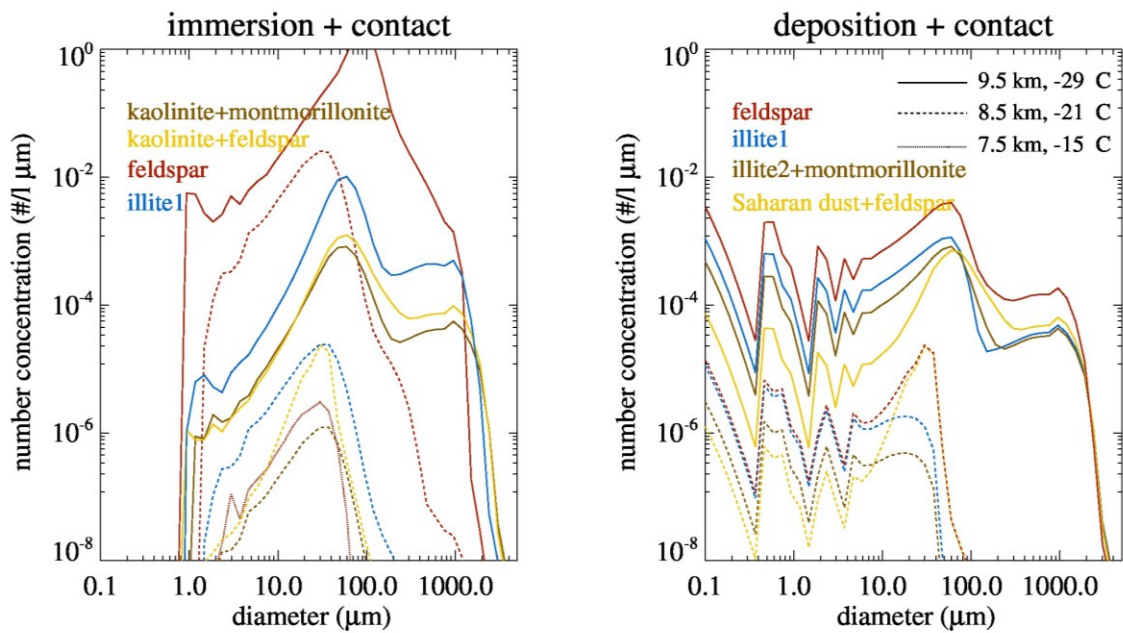
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11 Figure 10: Development of ice particle numbers with altitude and corresponding temperature,  
12 marked by different line styles, for deposition, contact and immersion freezing. Types of INP  
13 marked by colors. Model simulations with average continental number size distribution and  
14 with  $\Delta T = 2\text{K}$ . Number concentrations per l and  $\mu\text{m}$  as a function of particle diameter.

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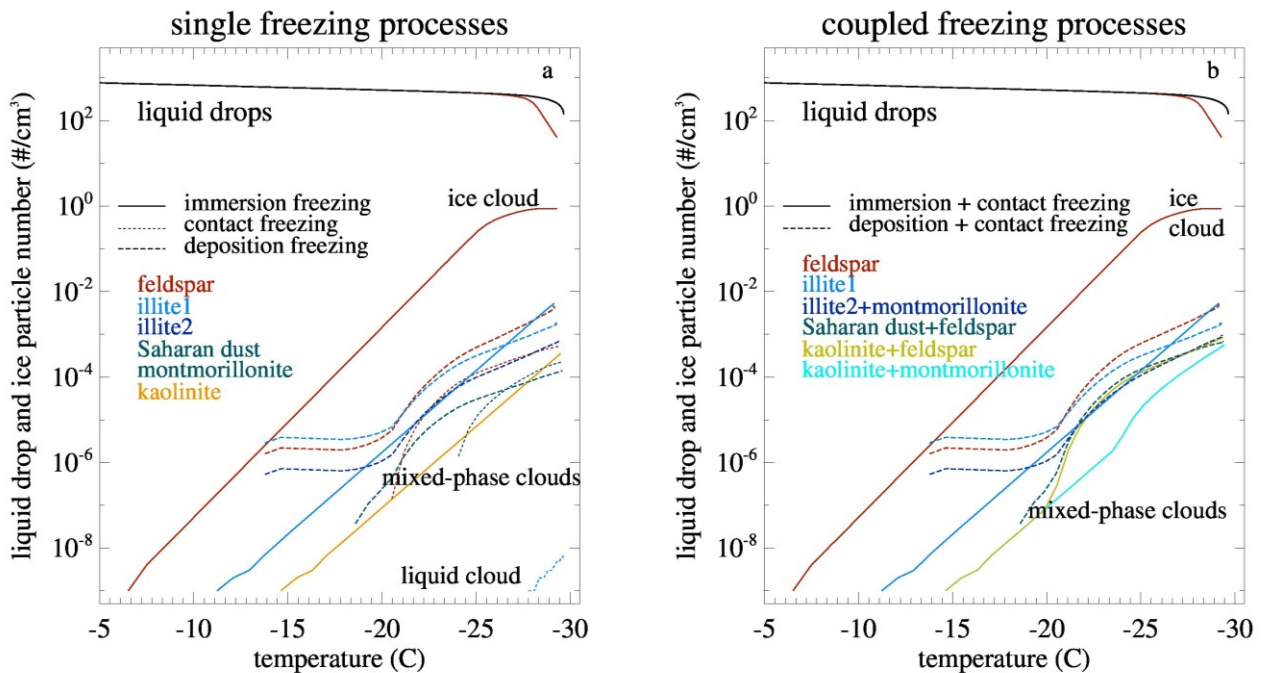
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 2 Figure 11: Development of ice particle numbers with altitude and corresponding temperature,  
 3 marked by different line styles, for coupled freezing processes. Types of INP marked by col-  
 4 colors. Model simulations with average continental number size distribution, and with  $\Delta T = 2K$ .  
 5 Number concentrations per l and  $\mu m$  as a function of particle diameter.

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 8 Figure 12: Total numbers of liquid drops and ice particles as a function of temperature for  
 9 single and coupled freezing processes which are marked by different line styles. Types of INP  
 10 marked by colors. Model simulations with average continental number size distribution and  
 11 with  $\Delta T = 2K$ . Numbers concentrations per  $cm^3$  as a function of temperature.