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_2 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, 1st 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 he 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 cm3'.

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