We thank Corinna Hoose for her careful review that revealed several sections in our manuscript that were insufficiently explained. The main changes in the revised manuscript concern
(i) the description of condensation and immersion freezing which is better clarified by addition of the corresponding model equations, and
(ii) the treatment of the 'deterministic' scheme that we now only apply to conditions where immersion freezing can occur, i.e. when conditions are supersaturated with respect to water. Since condensation freezing and immersion freezing can be both described by the CNT framework and depend on the surface properties of the IN (effective contact angle θ), we apply the contact angle distributions for the full temperature/supersaturation range that is covered in the parcel model simulations. All simulations using the deterministic scheme have been repeated and figures have been replaced.
All specific reviewer comments are addressed below in detail. We note that the equation numbers in the response refer to the new numbering in the revised manuscript.

Review by C. Hoose (Referee)

In this article, B. Ervens and G. Feingold investigate different parameterizations for immersion freezing, which are based on droplet freezing experiments by Lüönd et al. (2010). The five nucleation schemes are described clearly. As the parameterizations are all fitted to the data, they all do a reasonable job to reproduce them for the conditions under which the experiment was conducted. However, the authors show that they strongly diverge for different time scales, temperatures and particle sizes. Furthermore, the parameterizations have been implemented into an adiabatic parcel model, in which supersaturation varies during the ascent of the parcel and feeds back on ice nucleation.
This is a very timely study. Several different parameterization approaches of immersion freezing have been discussed in the recent literature, but so far, they have not been thoroughly compared with respect to their impact on ice formation in an ascending air parcel. I found the article very interesting and enlightening, but stumbled at a number of occasions. I hope that my comments, some of which are intended to provide more background on experimental findings on heterogeneous ice nucleation, help the authors to further improve this study. My main points are the following:

Reviewer comment: In this paper, N_{IN} and the term "IN number concentration" refer to the total number of kaolinite particles, and N_{ice} to the number of ice particles created through heterogeneous ice nucleation. This is different from the common terminology in most experimental studies, which use N_{IN} as the number of activated ice nuclei at a given temperature and relative humidity, i.e. a (usually small) subset of the kaolinite/dust/etc particles. Although this is rather a semantic point, I think it is important to clarify this because it can lead to a lot of confusion. I strongly recommend to adopt the common terminology and not to term all dust particles "IN".

Response: We agree with the reviewer that in the literature ambiguous definitions of 'ice nuclei' and 'ice crystal' concentrations are used and apologize that our definitions were not as clear and introduced some confusion.

Our definitions are based on the approach by Eidhammer et al. (2009) who pointed out that models using CNT need to constrain the number to particles that can freeze and they referred to these particles as 'ice nuclei' and to the number of frozen particles as 'ice crystals'. In our parcel model simulations, we assume that a population of internally-mixed ammonium sulfate/insoluble particles (N = 100 cm^{-3}). The exact choice of aerosol parameters is not of great importance to our discussion as we solely focus on feedbacks of the different nucleation schemes on cloud evolution.

We ascribe the IN properties of kaolinite as determined by Lüönd et al. (2010) to a small fraction of this aerosol population (4 L^{-1} / 100 cm^{-3} = 4e-5). In most of our conceptual simulations it is assumed that these particles have D_{IN} = 800 nm in order to remove the 'size aspect' of IN activation. We refer to these particles as 'potential ice nuclei (IN)'. We added the references as listed by the reviewer (introduction and Section 2.3) that show that our assumptions of the relatively low IN concentration and N_{IN}/N_{CCN} ration is in reasonably agreement with observations.

While in CFDC measurements all frozen particles are classified as 'ice nuclei', we would like to retain our different definitions of 'ice nuclei' and 'ice crystals' since in model approaches that take the time-dependence of freezing into account not all 'potential ice nuclei' freeze immediately and thus $N_{ice} \leq N_{IN}$. We added some references in the introduction that use similar terminology.

In the revised manuscript (Section 2.3.) we add more clearly our definitions of ice nuclei and ice crystals. In addition, we clarify that the predicted frozen fractions refer to the assumed number concentrations of IN, i.e. a fraction of 100% means that 4 L^{-1} (or 1 L^{-1}, in Figure 7 and 8d-f) particles are frozen, in approximate agreement with IN concentrations as determined from CFDC measurements.

Reviewer comment: My second point is actually related to the first one. For the parcel model studies, NIN (i.e. the kaolinite concentration) is prescribed to 4 l-1 (and 1 l-1 in two sensitivity experiments). No reference is given for these values, but my impression is that this choice is guided by typical atmospheric IN concentrations, as measured in a CFDC. However, it is important to note that atmospheric dust number concentrations, while very variable in space and time, are typically orders of magnitude higher! Measurements of dust number concentrations are usually only available in dust plumes and dust source regions, but see e.g. Penner et al. (2009), Fig. 5, or Hoose et al. (2010), Fig. 3, for simulated zonal average dust concentrations - even these are in the order of 1 cm-3. In my understanding, only a small fraction of the total dust concentration activates as ice nuclei in clouds, and the 100% activation (or frozen) fraction as displayed in Fig. 2 is normally not reached in the atmosphere.

Response: As pointed out in the response to the previous comment, we clarify that the frozen fraction always relates to the assumed number concentration of potential IN (4 or 1 L^{-1}, respectively). We thank the reviewer to pointing us to references that support the order of magnitude of our assumed particle concentrations and N_{IN}/N_{CCN} ratios that better justify our choices of these concentrations.

Reviewer comment: Therefore, probably the most interesting regime is outside the temperature range of Fig. 2, at temperatures above 241 K, where a small fraction of the kaolinite particles are active. (For example, Pinti et al. (2012) find much higher freezing onset points for bulk samples containing numerous kaolinite particles.)In contrast, more than 50% activation are reached in most simulations presented here. I recommend extending this study by using a higher dust number concentration as input to the adiabatic parcel model, and to focus on regimes (with a warmer start temperature for the parcel) where only a small fraction of the kaolinite particles activates to IN.

Response: We hope that our improved definitions above regarding our assumptions about number concentration of 'potential IN' clarify that the high predicted frozen fractions refer only to the values of N_{IN}. At higher temperatures the number concentration of frozen particles will be accordingly smaller resulting in a negligible IWC as compared to the much greater LWC. We have performed previously an extensive sensitivity study of the impact of IN concentrations in different temperature regimes and have shown that for the conditions considered in the parcel model (e.g. no particle removal by precipitation) glaciation of clouds might occur if $N_{IN} > \sim 5\ L^{-1}$ (Ervens et al., 2011).

Reviewer comment: The description of the deterministic scheme should be improved, some of the statements are inaccurate. See detailed comments below.

Response: We used the deterministic approach as suggested in the experimental study (Lüönd et al. (2010)) that forms the basis for most of our simulations. We make this fact clearer throughout the revised manuscript (Sections 2.1.5, 2.3, 4.1.2) and point out that we have redone all simulations using the deterministic approach by only allowing ice nucleation if $S_w > 1$ in order to account for the dependence of condensation freezing on supersaturation which is not covered by the approach by Lüönd et al. (2010) that was derived under supersaturated conditions. We will give more detailed responses to the specific comments below.

Reviewer comment: It is stated in the abstract that laboratory experiments "often" report a time dependent behaviour of ice nucleation. This is not true; on the contrary, of the few experimental setups which actually were able to investigate the time dependence of ice nucleation (DeMott, 1990; Niedermeier et al., 2010; Murray et al., 2010; Broadley et al., 2011), only a small number actually found an increase of the activated fraction with time (or at slower cooling rates).

Response: We removed the 'often' in the abstract in order to avoid the impression that indeed a majority of laboratory studies find time-dependent freezing processes. We reworded the introduction and discuss specific studies such as the ones mentioned by the reviewer that discuss the stochastic nature of the freezing process and that find an increase in frozen fraction as a function of cooling rate.

Detailed comments

Reviewer comment: Section 2.1.1: Please give more details on how the prefactor in eq. (1), F_{act} and r_{germ} are parameterized (possibly in an appendix), because different formulations can lead to very different results.

Response: The expressions of the prefactor, F_{act} and r_{germ} were taken from the framework by Khvorostyanov and Curry (2004).

We changed Eq-1 and include now the prefactor explicitly

$$J = \frac{kT}{h}\frac{\pi}{6} \cdot 10^{19}\, m^{-2}\, D_{IN}^{2}\, exp\left[-\frac{\Delta F_{act} - \Delta\, F_{cr}}{kT}\right] \tag{1}$$

We add the equation for ΔF_{act} that is based on Jeffery and Austin (1997) and also used in the framework by Khvorostyanov and Curry (2004); (2005)

$$\Delta F_{act} = 0.694 \cdot 10^{-12} \cdot (1.+0.0027 \cdot (T_C\ [^{\circ}C] +30)) \tag{2}$$

Since both reviewers asked for more information on our description of condensation and immersion freezing, we also add the equations for the germ radius in detail, following the approach of Khvorostyanov and Curry (2004).

For immersion freezing, we use

$$r_{germ} = \frac{2\sigma_{is}}{\rho_{ice} L_m^{ef} ln\left(\frac{T_0}{T}\right) + \frac{RT\rho_{ice}}{M_w} H_c - \frac{2\sigma_{sa}}{r_d}} \tag{4}$$

with

$$H_c = \frac{2\sigma_{sa} M_w}{\rho_w RT\, r_d} - \frac{\nu\Phi(1-\varepsilon_{insol}) M_w \rho_s r_s^3}{M_s \rho_w (r_s^3 - r_d^3)} \tag{5}$$

Since particles at $S_w < 1$ are near equilibrium

$$\ln S_w = H_c \tag{6}$$

for deliquesced particles, it is assumed

$$r_{germ} = \frac{2\sigma_{is}}{\rho_{ice} L_m^{ef} ln\left(\frac{T_0}{T}\right) S_w^{\left(\frac{RT}{M_w L_m^{ef}}\right)} - \frac{2\sigma_{sa}}{r_d}} \tag{7}$$

The definitions of all parameters are added in the manuscript (Section 2.1.1).

Reviewer comment: `Section 2.1.3: It would be helpful to include information on in how far the "internally mixed soccer ball scheme" is equivalent to the "active site distribution" by Marcolli et al. (2007) and the "Nucleation Probability Dispersion Function" by Barahona (2012).`

Response: The "internally mixed soccer ball scheme" represents a simplified scheme as compared to the schemes the reviewers lists here (and similar ones).

These surface-area-based approaches assume the distribution of nucleation sites over the total surface of an aerosol population, in order to represent the random distribution of such sites (that are characterized by contact angles or specific freezing temperatures, i.e. 'active sites') on a sample of aerosol particles. This randomness leads to different distributions of nucleation sites on each particle.

Our "internally-mixed soccer ball scheme" is a hypothetical intermediate scheme between the 1θ scheme where all particles are the identical with a single θ and the 'externally mixed soccer ball scheme' that represents distributions of nucleation sites as assumed by Marcolli et al. (2007), Barahona (2012) and others. We introduced this additional scheme, in order to increase the complexity of the nucleation scheme in a stepwise manner which allows us to systematically explore the effects of contact angle distributions. We discuss now these similarities and differences in Section 2.1.

Reviewer comment: `Section 2.1.5: The deterministic scheme, as given by equation (6), is often called "active site density scheme" or "ns-scheme", and these names should show up here. I find it confusing that Fletcher, Meyers and DeMott et al. (2010) are given as prominent examples - although they are of course deterministic scheme, they follow a completely different approach. Closer to what is actually implemented here are Connolly et al. (2009) and Niedermeier et al. (2010). These do not suffer from any missing constraints to the total IN (or dust) concentration. Neither does Phillips et al. (2008), by the way.`

Response: We extended this paragraph and describe in a more systematic way the different approaches that are used to parameterize the number concentration of ice particles. As suggested by the reviewer, we distinguish more carefully between

(i) laboratory based expressions that report the integrated number concentrations of frozen particles as a function of temperature and/or supersaturation (i.e. using k(T) parameter; Eq-11)

(ii) expressions by De Mott et al (2010) etc that are based on observations. Such empirically-derived expressions are often used in models since they were derived from observations and thus might represent best the variability in the atmosphere but suffer from a poor constraint on the upper limit if N_{IN} and the lack of the sensitivities of the underlying physical processes.

Reviewer comment: `Section 2.3 More information is needed on how condensation freezing is treated. Is the freezing point depression taken into account? How is this done? What happens to evaporating droplets? Are efflorescence and deflorescence treated explicitly?`

Response: The equation set by Khvorostyanov and Curry (2004) adopted here (Eq. 1-7) explicitly considers the drop radius and solute concentration. Thus, freezing point depression is included in our model.
Our simulations are restricted to a single ascent of an air parcel. Even under conditions when the parcel becomes subsaturated with respect to water, S_w is still above ~80% (Figure 5), i.e. way above the efflorescence point of ammonium sulfate particles. Thus, all particles that are not activated into cloud droplets are always in a deliquesced state throughout the simulations. We have added text in Section 2.3. to clarify this.

Reviewer comment: Section 3.3 and also in the figure captions: I don't think the variation of Sice needs to be mentioned, as this is not an independent variable. Swat is kept constant.

Response: We agree that the temperature determines S_{ice} at a fixed S_w and thus the first two sentences in Section 3.3. are trivial. We simplified them by removing the S_{ice} range. However, we kept the S_{ice} information in Figure 3b in order to provide some information on S_{ice} values and sensitivities to readers who are not fully familiar with the T/S_{ice} space.

Reviewer comment: page 7181, line 13: "since the supersaturation is sufficiently high": I would argue that it would be better to say that the temperature is sufficiently low. For immersion freezing, J is primarily a function of T and only indirectly a function of S_{ice}. Similar formulations are also found in section 4.1.2.

Response: While in equilibrium temperature and supersaturation are unambiguously linked, in the non-equilibrium states as encountered in clouds, supersaturation is not only affected by temperature. In a hypothetical state where the temperature is sufficiently low but the water vapor is efficiently depleted by ice growth, no efficient ice nucleation will occur.
The temperature is not significantly affected by ice formation and growth; however, as shown in Figures 4e and 5, the supersaturation might decline at a sufficiently high concentration of ice particles. Under such conditions, further ice nucleation is prevented which is reflected by the vertical lines in Figure 4e.
At cloud base (referred to on p. 7181 in the ACPD manuscript) such competition effects are not effective and a sufficiently high reservoir of water vapor is available to allow the direct link between temperature and supersaturation. In order to clarify this link we change the sentence in Section 4.1.1 to
'… near cloud base where the low temperature and sufficiently high supply of water vapor allow ice nucleation of IN with relatively low θ'.
In Section 4.1.2., we clarify the feedbacks of microphysical processes (ice nucleation and growth) on the water vapor supply and point out that despite sufficiently low temperature, ice nucleation by condensation freezing does not occur since S_{ice} is too low.

Reviewer comment: page 7186, line 5: "the lack of comprehensive parameter sets ... at 253K< T <263K": This criticism should be formulated more carefully: Which parameters are required? After all, there are numerous laboratory studies which cover this temperature range. Most recent examples are Pinti et al. (2012) and Niemand et al. (2012).

Response: It was not our intention to ignore the numerous laboratory studies that were performed in this temperature range. What we meant to say, and now clarify, is that a consistent parameter set as listed in Table 2 for all five schemes from a single experimental study for kaolinite (T ~ 240 K) is not available for IN at higher temperatures.

The reviewer is right that many data are available and we refer to them in the revised manuscript (Section 4.1.3). We add that the spread in the onset of freezing is at the lower end of observed values (e.g. Pinti et al. (2012)) which suggests that the variability of IN surface properties tends to be greater at higher temperatures. Note that we arbitrarily assumed here the same width of the θPDF as for kaolinite by Lüönd et al. (2010).

In the exploratory model simulations, we have chosen 'ideal' temperature ranges that show the opposite trend in terms of T-Γ relationships in order to highlight the potential combined effects of nucleation scheme and initial inherent growth ratios. Different initial temperatures and temperature ranges might translate into different absolute effects; however, the qualitative findings from our model studies will be unchanged.

Reviewer comment: page 7188, line 20: It is unclear to me why the size dependency should be stronger for the deterministic approach than for the other schemes. Aren't they all roughly proportional to surface area? Is the size dependency of f(m,x) already relevant for diameters around 800 nm? Please explain.

Response: In the deterministic approach as used here, the surface area is only in the exponent and thus a direct relationship between S_{IN} and F_{Fr} can be seen. In CNT, J depends on D_{IN} in a more complex manner since it is not only dependent on D_{IN}^2 (Eq-1) but also included in the calculation of the germ radius (m_s, r_s, ε_{insol}).

Reviewer comment: Fig.2: Please include Lüönd et al. (2010)'s data points as a reference.

Response: We added Lüönd et al.'s experimental data (without error bars) to Figure 2.

Reviewer comment: Fig.3: Why are the dotted curves not smooth?

Response: In the original simulations, we calculated F_{fr} for a rather coarse time resolution (dt = 1, 5, 10 s… etc). We redid the simulations with a higher time resolution. This improvement removed the discontinuities. We replaced Figure 3.

Reviewer comment: `Fig.4: Why are there steps in the ice crystal number concentration, e.g. in the yellow curve in 4(b)? If this is due to the discretization of the contact angle spectrum, then more bins would be needed.`

Response: CNT predicts that the nucleation of a (small) number of IN at any temperature a at every model time step (1 second). In theory, this would lead to a new ice class from each IN size class (*i*) every second in the model simulations, which would result in e.g., to up to 3000 ice classes in the w = 10 cm/s simulations (300 m / 10 cm/s / 1 s) if we assume a monodisperse, internally-mixed IN distribution. This number multiplies with the number of 'externally mixed' particles (20) in the soccer(ext) scheme and the θPDF scheme and with the number of IN size classes (1 for most simulations, 10 for simulations in Figure 8), i.e. $i \leq 600{,}000$.

In order to keep *i* computationally manageable throughout the simulations, we use 'probability steps', i.e. we only fill a new ice class if 2.5% of an IN size class and particle type of this size (in the externally mixed schemes) is predicted to freeze. While for the 1θ or the soccer(int) scheme, it might be feasible to use a finer resolution, we have chosen the same steps for all simulations for the sake of consistency.

The advantage of this method as compared to other methods that combine ice particles of similar sizes into bins is that we can always track every individual ice particles back to its original aerosol particle and its properties (size, contact angles …) whereas this information is lost in the usual binning methods.

The fact that the results of the 1θ scheme also exhibit such steps indeed shows that it is not a poor resolution of the contact angle distributions but due to the 'probability steps'. We have explained this method in more detail in our previous study Ervens et al. (2011) and briefly in Section 2.3. We also added this explanation in the caption of Figure 4 in order to avoid the impression that a poor resolution of contact angles might cause these steps.

Reviewer comment: `Fig.4(f): For the deterministic model, why does Nice still increase although all liquid droplets seem to have evaporated? Even though eq.(6) doesn't include any explicit dependency on S, it can only be applied as long as the kaolinite particles are immersed in water.`

Response: In our updated simulations, we excluded any ice nucleation by the deterministic scheme if $S_w < 1$ recognizing the fact that condensation freezing has to include supersaturation as a key parameter. In the original simulations, we had applied this equation over the complete T range without considering S_w. We realized that this approach is oversimplified and unjustified and, thus, we have changed all figures (4, 5, 7, 8, 9) where parcel model results from the deterministic approach are presented. Since we start our simulations near cloud base ($S_w = 0.99$), the evolution of N_{ice} from the deterministic scheme is only marginally different. Largest differences are indeed seen in Figure 4e where ice nucleation stops upon the onset the Bergeron-Findeisen-Process.

In Section 2.1.5, we explain now Eq-12 is more appropriate if $S_w > 1$. The other schemes that are all based on CNT are still applied over the whole temperature range since CNT describes ice formation on nucleation sites (characterized by an effective contact angle) in any temperature and supersaturation regime.

Reviewer comment: Table 1: The choices with respect to number of nucleation sites/particle and number of different particles should be commented on in the text.

Response: This information is now included in Section 2.2.

Technical comments

- page 7172, lines 1 and 5: please give the values in SI units.

We changed the units of surface tension and the Boltzmann constant accordingly.

- page 7176, line 14: φ- > Φ

We will make sure that in the copyedited version of the manuscript these symbols are shown correctly.

- eq.9: i- > j

We changed this symbol.

- Fig.4: It would be helpful to include a vertical temperature axis similar to Fig. 7.

We added the temperature scale to the figures that show IWC (Figures 4b, d, f).

- Table 1: b1 seems to be missing in the column heading. I can’t identify what footnote 2 refers to.

The ‘b’ in the numerator of the exponential term should have been b1. This omission was introduced during the copyediting of the manuscript.
In addition, we replaced the apparent exponent 2 in the same equation by (*2) in order to make clear that it is not an exponent but refers to the footnote.

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