Response to Referee #2 (RC C486)

2.1 Page1401, line14: Deposition nucleation is not restricted to water sub-saturated conditions, this could be made clearer.

The sentence now reads

‘deposition mode occurs under ice supersaturated conditions via deposition of water vapour onto the INP surface without the formation of bulk liquid water’

2.2 Page 1401, line16-21: Mixed-phase clouds always contain water droplets; this is not a sufficient argument to exclude deposition nucleation. Try to clarify and reformulate.

The paragraph now reads

‘Observational studies show strong evidence that above homogeneous freezing temperatures the formation of ice is commonly preceded by the activation of the liquid phase, hence the glaciation of an air parcel transitions through a mixed-phase regime (Ansmann et al., 2009; de Boer et al., 2011; Field et al., 2012; Westbrook and Illingworth, 2013). Ansmann et al., 2009 found that in 99 % of cases the production of ice occurred after the formation of a liquid phase, and similarly, de Boer et al., 2011 found that air parcels under ice supersaturated conditions did not produce ice until after a liquid layer was formed. This suggests that deposition and condensation mode ice nucleation play a secondary role in the glaciation of these clouds’

2.3 Page1401, line 24: Consider adding Durant and Shaw (2005) to the list of references for evaporation freezing.

The reference has been added as suggested.

2.4 Page 1402, line 7: Consider adding an example to illustrate the range of ice nucleation ability.

The following sentence has been added:

‘For example, bacteria INP species belonging to the *Pseudomonas* genera catalyse freezing at temperatures above 265 K and exhibit a steep function of freezing rate (Wolber et al., 1986; Mortazavi et al., 2008), whereas mineral dust has been found to catalyse freezing at lower temperatures and exhibit a weaker gradient (Niedermeier et al., 2010).’

2.5 Page 1403, line 6: Can you add a statement for what kind of materials the stochastic nature is more important?

The following sentence has been added:

'Wright et al. (2013) tested the cooling-rate dependence for a range of INP species and found variability in their cooling-rate dependence. For the minerals kaolinite, and montmorillonite, along with flame soot, the median freezing temperature of a droplet population decreased by ~3 K upon a factor of ~100 increase in cooling rate; conversely, the bacteria species showed no change for the same increase.'

Additionally, the study now includes a normalised comparison of the variability in the stochastic nature of atmospherically relevant INPs from existing literature. The data, determined using the FROST framework, is presented in Table 2 and also shown in Fig. 11.

2.6 Page 1403, line 13: Name the properties that have been tested. Can you think of other possible, but not tested relationships?

This refers to the paragraph: 'In this study we use a multiple component stochastic model to establish the key relationships between the physical properties of an INP and its observable time-dependent behaviour, which are then captured in a simple framework.'

In this study we have found evidence that the nucleation rate coefficient solely determines the time-dependent behaviour. The 'physical properties' refer to the INP characteristics / physiochemical properties that result in the gradient of the nucleation rate coefficient, and therefore the observed time-dependence. As we do not know how these physiochemical properties determine the gradient, 'physical properties' will be replaced with 'nucleation rate coefficient' in the revised manuscript.

2.7 Page 1404, line 5-6: Please name the materials you are referring to.

The following has been added to the sentence:

'….including the mineral kaolinite (Murray et al., 2011) and silver iodide (Heneghan et al., 2001).'

2.8 Page1407, line 7: Based on the reasons for inter-particle variability in ice nucleation ability given on page 404, can you justify the assumption of a Gaussian distribution?

The following sentence has been added to the paragraph:

'Although there is evidence for multiple components the distribution of such components is not currently known and difficult to infer. For simplicity a Gaussian distribution was used following previous studies (Niedermeier et al., 2010; Broadley et al., 2012; Wright and Petters, 2013)'

2.9 Page 1419, line 2-4: What is the RMSE of this fit?

The sentence was amended as follows:

'The best fit was determined by minimisation of the root-mean-square-error (RMSE) between the data and a linear fit to ln(n_s) for data where $T_{experiment} \leq 262.65$ K (-10.5 °C); this temperature was chosen to limit effects from anomalous high temperature freezing events. This fitting procedure, with a RMSE value of 0.009, resulted in $\lambda = 3.4$ K^{-1} and is shown in Fig. 6b'

2.10 Page 1421, line 17: Hoyle et al., 2011 used a fluidized bed aerosol generator with an upper size cut-off at 3μm for the ZINC experiment not 300 μm. However the ln(n_s) values in Fig. 9.a) seem to be correct. Hoyle et al. reported additional DSC data. Consider including this measurements to your dataset to demonstrate the applicability of FROST to reconcile data from three different experiments.

The stated size range has been corrected; the data is not affected.

Unfortunately, we were not able to include the Hoyle et al. (2011) DSC data in this study. The FROST framework was developed for application to quantitative data from cumulative fraction frozen data for a narrow size range of droplets. No fraction frozen curves are reported and it is therefore not possible to use FROST.

2.11 Page 1423, line 11-23: It seems that more efficient IN generally exhibit a larger λ and therefore a weaker time-dependence. Can you comment on that?

As observed by the reviewer the time-dependent behaviour of the tested species appears to correlate with their relative nucleating efficiency. For example, bacteria are often very efficient INP species and mineral dusts are generally less efficient (Murray et al., 2012; Hoose and Möhler, 2012).

We have added the following discussion in section 5.1:

'It is also apparent from Fig. 11 that more efficient INPs tend to exhibit a larger value of λ. This behaviour was also noted by Vali (2014). For example, bacterial INPs and soils which contain some of the most efficient INPs we know of also have the largest values of λ. Interestingly, classical nucleation theory predicts that λ is larger at higher temperatures. However, there are also exceptions to this 'rule'. Values of λ determined from Fornea et al. (2009) for peat and volcanic ash are very similar, but the peat sample nucleated ice at much warmer temperatures. It is also interesting to note that in many previous studies examining the role of time dependence (Vali, 2008; Vali and Stansbury, 1966; Vali, 1994; Welti et al., 2012), which formed the basis of the argument that time dependence is of secondary importance, the materials used have larger λ values and therefore less sensitive to temporal conditions. More work needs to be done on what factors control the value of λ.'

2.12 Page 1414, line 14: Check the numbers of equations you refer to.

Done, and amended.

2.13 Page 1419, line 21: remove "K" after $\lambda = -3.4$

We have corrected the units of λ throughout the paper. It should be K^{-1}.

2.14 Page 1419, line 22: σ,μ are inverted

Corrected.

2.15 Fig. 5, 6, and 8: Consider homogenizing this figures in terms of what is shown in which column. Additionally it would be interesting to see F, n_s and J for all three datasets.

An excellent suggestion. Data for all examples have been presented in a homogenised manner as suggested by the reviewer. We have also adjusted the text where necessary in order to reflect these changes.

2.16 It might be interesting to discuss the influence of particle size distribution and surface area immersed in a droplet. Can the PICOLITRE experiments on KGa-1b and feldspar or the experiments on larger K-SA particles be reconciled with the data shown in Fig. 5, 6 and 8?

The K-feldspar sample used in this study was also used in Atkinson et al. (2013); in the study the surface area of sample per droplet was increased by several orders of magnitude and once normalised to surface area the data fell onto a single line over all temperatures tested. The fit to the K-feldspar data from Atkinson et al. (2013) will be included in Fig 6 and a paragraph will be added to briefly discuss the dependence on surface area, and reads as follows:

'Figure 6e also includes the fit to K-feldspar data presented in Atkinson et al. (2013). In their study the surface area of K-feldspar per droplet was increased by two orders of magnitude to examine the dependence of freezing rate on surface area and all experiments were performed at a cooling rate of 1 K min^{-1}. The parameterisation from Atkinson et al. (2013), based on data with variable surface areas, is in good agreement with data from this study.'

In addition, we have also included a discussion of the kaolinite (KGa-1b) data from Murray et al. (2011) in section 4.1 in response to Referee #1 which highlights the agreement between different experiments with different numbers of particles (and different surface areas) per droplet.

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