Author's Response to reviews

Dear Dr Petters,

The following text details the point-by-point responses to the comments from referees, and the subsequent changes that have been made, where appropriate, to the manuscript; these are a reproduction of the responses to the referee comments as shown on ACPD. Our response to suggestions by the editor follows.

As you are likely aware we have had extensive communication with Gabor Vali in order to produce a consistent set of nomenclature between this paper and Gabor's paper recently published in ACP (Vali, 2014). It is hoped that the simultaneous presentation and use of this nomenclature will promote consistency within the community. In light of this shared nomenclature it was necessary to make some minor changes to the derivation of the framework and subsequent references throughout the manuscript. Briefly, A has replaced s as the surface area (cm^2), $J_{apparent}$ has been replaced with R/A where R is the freezing rate (s^{-1}), ω has been introduced to describe the temperature dependence of R ($-d\ln(R/A)/dT$), and in order to make a comparison between ω and λ the sign of λ has been changed so that $\lambda = -d\ln(J)/dT$. These changes are reflected throughout the manuscript and Supplement. Additionally, IN (ice nuclei) has been replaced with INP (ice nucleating particle), and J_s has replaced J.

Response to Referee #1 (RC C390)

> *1.1. Page 1401: You write that contact ice nucleation occur "through the collision between an IN and the air–liquid interface of a supercooled droplet". You may include that there are two different possibilities: outside-in and inside-out collision. To my understanding the latter mechanism is the important one for freezing of evaporating supercooled droplets (as you mention in line 22-23)*

The sentence now reads

'*contact freezing* through an outside-in or inside-out contact between an INP and the air-liquid interface of a supercooled droplet'

1.2. Page 1402: Ervens and Feingold (2013) show in their study (single component system, based on CNT i.e., stochastic approach) that a change in temperature has a larger effect on the frozen fraction compared to a change in time (one to several orders of magnitude depending on the chosen contact angle). But this finding is not in contrast to the findings of Kulkarni and Dobbie (2010), Murray et al. (2010) and Welti et al. (2012). An isothermal experiment only shows the stochastic nature of freezing, i.e., the time dependence. But the interesting question is how does a change in temperature relate to a change in time. For example, Welti et al. (2012) showed for their kaolinite sample that a change of T =1K in temperature relates to a time ratio change (t2/t1) of a factor 10, i.e., temperature is more important than time. Please rephrase the paragraph accordingly.

We did not intend to imply that Ervens and Feingold's paper was 'in contrast' to other studies of time dependence. We have removed the term 'in contrast' and amended the section which now reads:

'Similarly, Ervens and Feingold (2013) recently performed a sensitivity study which highlighted changes in temperature as being the most important factor in droplet freezing sensitivity. Nevertheless, a number of studies show that there is a sensitivity of ice nucleation to time. For example….'

1.3. Page 1403: The probability for critical cluster formation depends on droplet volume and time, but most important on temperature! The lower T, the higher is the freezing probability.

Sentence rewritten to read as follows:

'For homogeneous nucleation, the probability of a critical cluster forming rapidly increases with decreasing temperature (Stan et al., 2009; Murray et al., 2010). Additionally, the probability is increased for both larger droplet volumes and longer time scales.'

1.4. Page 1406, Eq. 1: Do you consider that after each time step the liquid droplet number has to be adjusted so that the total number (frozen plus liquid droplets) keeps constant?

Yes, so that $n_{\mathrm{liquid}\,i+1} = n_{\mathrm{liquid},i} - n_{\mathrm{frozen},i}$. This has been made clearer in the manuscript by adding the following:

'Upon subsequent steps the number of available droplets is adjusted so that $n_{\mathrm{liquid}\,i+1} = n_{\mathrm{liquid},i} - n_{\mathrm{frozen},i}$.'

1.5. Maybe I missed that piece of information: How many droplets do you examine in your cold stage cell studies for kaolinite and feldspar. What are the measurement uncertainties for the determined frozen fractions due to temperature uncertainty (0.4 K for isothermal experiments), considered droplet number (Poisson statistics), etc.?

As stated in the text (page 1416 line 8; page 1418 line 9) ~40 droplets were typically used in the microlitre experiments. Uncertainties in measuring the fraction frozen are solely due to the uncertainty in the measured temperature profile (estimated by Whale et al. (2014) as ± 0.4 K), arising from the temperature probe and the observed range in melting temperatures.

The uncertainties in measurements are stated in figure captions but not in the main body of the paper. Sentences in sections 4.1 and 4.2 have been added to state these uncertainties and their origins:

'All experiments were performed with a weight fraction of 0.01, corresponding to a surface area of 1.178 ± 0.3 cm^2 calculated using a specific surface area of 11.8 ± 0.8 m^2 g^{-1} (Murray et al., 2011). The uncertainty in surface area per droplet primarily arises from uncertainty in specific surface area measurements and droplet volume. The temperature uncertainty, arising from the temperature probe and observed range in melting temperatures, has been estimated by Whale et al. (2014) as ± 0.4 K.'

and

'Each droplet contained a weight fraction 0.001 of K-feldspar, corresponding to a surface area of $1.85 \times 10^{-2} \pm 0.004$ cm^2 calculated using a specific surface area of 1.86 m^2 g^{-1} (Whale et al., 2014).'

> *1.6. Chapter 4.1 and Fig. 4 and 5: Regarding the microlitre data in Fig. 4a, I would think it shows a less steep slope compared to the picolitre data. Is there maybe an obvious difference between the two samples? And why does the frozen fraction in Fig. 5a for 0.5 K/min and 1K/min cooling rate does not reach 1?*

The microlitre data may have a slightly less steep slope than the picolitre data, but it is not clear if this is significant given the scatter in the experimental data. Also note that the expected isothermal decay (Fig 4b) shows very good agreement when the slope fit to the combined dataset (both microlitre and picolitre stages) is used, suggesting that there is no significant difference between the samples used in the two sets of experiments.

The fraction frozen does not reach unity in the faster cooling rate experiments above the temperature at which pure water droplets begin to freeze: $\sim$-20.5 °C (252.65 K). However, we did notice that we had cut some of this data off at -20°C rather than -20.5°C and this has been corrected. This resulted in small changes to the associated RMSE values but did not affect the results.

> *1.7. Fig. 8: Please include the measurement uncertainties also in the raw data. For me it looks like that the raw data already agrees within the uncertainties if the error bars from Fig. 8b) and 8d) are used.*

The error bars have been included. The RMSE values decrease upon the application of FROST, suggesting that the modification increases the agreement.

> *1.8. Can you also plot a figure for the Welti data as you did in Fig. 4b and Fig. 7, i.e., calculating μ and for this kaolinite sample?*

A new figure (Fig. 9) has been included that shows the WELTI data plotted as isothermals, with the expected decay included as dashed lines. It was not necessary to use the MCSM to do this as there was no cooling prior to the isothermal and the original n_s function could be used. A new paragraph has been included in the section to discuss the figure and the subsequent findings and reads as follows:

'Similar to the kaolinite and K-feldspar examples the determined value of λ was used to reproduce the expected decay of liquid droplets over time. With CFDC instruments the cooling from ambient temperature to the experimental temperature is very rapid and therefore the distribution of INP efficiency per droplet can be assumed to be represented by the function of $n_s(T')$ determined in Fig. 8e. To calculate the expected decay of liquid droplets with time Eq. (26) was used with the value of λ (2.19 K^{-1}) determined previously. The experimental data, along with the expected decay, is shown in Fig. 9. It can be seen that at high temperatures (241 to 239 K) the FROST framework is able to reproduce the experimental decay very well. However, at lower temperatures (238 to 236 K) there are large differences, especially for longer residence times. The reported errors bars are large for the lowest temperature data and suggest an increasing uncertainty with decreasing temperature. Also the fraction of droplets frozen is expected to increase with decreasing temperature as stated by WELTI. This suggests a potential experimental issue, which would explain the discrepancies.'

> *1.9. Looking separately on Fig. 9a, I would think that both data sets are clearly distinguishable through different slopes or cannot completely be represented by a single straight line curve (i.e., your straight line fit cannot be extrapolated to higher T). So, I am not completely convinced that the volcanic ash sample behaves like a single component system (Can you please plot the nucleation rates here? If it behaves like single-component, the rates for both experiments should fall onto one line without fitting.). As already stated in the text, more isothermal or cooling rate experiments would be needed to prove this assumption.*

It is worth noting that a single component system does not necessarily have to be characterised by a linear function of $\ln(n_s)$ vs T. The same would apply for such functions as derived by CNT. The FROST framework would still work but would simply require the use of a temperature dependent value of λ. However, this still needs to be confirmed with data and extensions of the existing derivations. For this reason a linear fit was used for this dataset.

The figure now includes data on derived R/A values (~ nucleation rate) as suggested by the reviewer – the data falls onto a single line which provides additional evidence of the volcanic ash sample behaving as a single-component system. The gradient of the fit to the combined dataset (ω = 0.55 K^{-1}) is similar to the minimised value of λ (0.60 K^{-1}). The $\ln(n_s)$ RMSE value determined from a linear fit to each modified dataset is 0.25 using λ, and 0.26 using ω. This suggests that the volcanic ash sample is a single-component system and therefore $\lambda = \omega$. However, uncertainty in these data arises from the assumed surface-area per droplet originally determined by Murray et al. (2012) to derive $n_s(T)$ from the $f(T)$ data.

We also now include experimental observations from Fornea et al. (2009) using a volcanic ash sample from Mount St. Helens and use FROST to determine λ. The experiments included cooling and freeze-thaw experiments. To determine a value of λ from the freeze-thaw experiments an empirical relationship between λ and the variability in freezing temperature $\sigma_{T\mathrm{freeze}}$ was determined using the MCSM model. The relationship is additionally included in the framework development section. The relationship bears a significant resemblance to that presented in Wright and Petters (2013), with the minor differences attributed to the methods used in determining the relationship. This relationship is introduced in the new section 3.3. Applying FROST to the FORNEA data results in λ = 0.64 K^{-1} for both experimental methods. The consistency in λ values between experimental methods and volcanic ash samples supports the conclusion that volcanic ash behaves as a single component species. The new analysis of the Fornea data is discussed in the last two paragraphs of section 4.4.

*1.10.	Page 1401, line 9: Replace warmer temperature through higher temperature.*

Done.

*1.11.	Page 1409, line 1: It should read n_frozen/n_liquid. However, to my understanding the frozen fraction is defined as the frozen droplet number divided through the total number of particles (i.e, sum of frozen and liquid droplets). I am just wondering about the definition of Eq. (13), is it valid with your fraction frozen definition?*

For these derivations n_{liquid} refers to the initial number of liquid droplets. This should be N_{liquid} and has been amended. The subsequent derivations are not affected.

*1.12.	Page 1412, line 16: I am not sure about the sign for the cooling rate: Is it positive or negative? Intuitively I would think r_cool > 0 K/min?*

A positive cooling rate implies a decrease in temperature, and therefore on this occasion was incorrect. This has been corrected and clarified throughout the revised manuscript.

*1.13.	Page 1419, line 1: Please add that T' = T_exp – (r)*

Done.

*1.14.	As you investigated droplets featuring more than one particle you should avoid saying that kaolinite is a uniform species since you can not prove it. Due to a high amount of particles per droplet it might be possible that there is a high probability for droplets featuring particles with similar ice nucleation activity. Welti et al. (2012) also used kaolinite, indeed they used a sample from Fluka which differs from your sample. However, they show that there is a particle-to-particle variability for their kaolinite sample. In my opinion you should only state that for your investigations the kaolinite sample can be treated as a single component system but there could be multiple IN populations leading to different results if droplets with single particles are regarded (you have this kind of interpretation on page 1417/1418).*

This is a very sensible comment which should be addressed in our paper. We have included a discussion around this point at the end of section 4.1:

'While nucleation by this kaolinite sample can be treated as a single component, this does not necessarily mean that this sample is uniform (i.e., there is no inter-particle variability) because there are many particles per droplet in the experiment. It is possible, but unlikely, that droplets contain a distribution of particles with diverse ice nucleating abilities, but where freezing in all droplets happens to be controlled by particles with similar ice nucleating activity. This is very unlikely given that the number of kaolinite particles in the PICOLITRE experiments ranges from just a few 10s to tens of thousands and all produce consistent values of J_s (Murray et al., 2011). In contrast, the Fluka kaolinite sample used by Welti et al. (2012), which is known to contain particles of very efficient feldspar (Atkinson et al., 2013), is a diverse species (as will be demonstrated in Sect. 4.3).

In summary, kaolinite KGa1b from the clay mineral society is an example of a material which most likely has uniform ice nucleating properties and can be described with a single-component stochastic model.'

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 $T_{experiment}$ ≤ 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 λ = 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 Mohler, 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 λ = −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.

Response to suggestion by editor

Following the suggestions by the editor the revised manuscript now considers and incorporates the observations from the study by Wright et al. (2013) in which the time-dependent freezing characteristics of a wide range of atmospherically relevant INPs were experimentally determined. References to the method and results have been incorporated into the revised manuscript throughout. The results from Wright et al. (2013) have been applied to the FROST framework and

subsequent λ values are included in Fig. 11 along with those determined from other studies in the literature. The results are incorporated into a discussion on the observed range in λ in Sect. 5.1.

Regards,

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