Authors' responses to comments by Referee #1

We thank Anonymous Referee #1 for his/her thoughtful comments and useful discussion. Below are our point-by-point responses.

Reviewer's comment [1]:

My main concern with the paper is the inclusion of a 'correction factor' (cf) in eq 2. I am concerned that the validity of the approach outlined by the authors in this and previous papers is undermined by the inclusion of what could be interpreted as a 'fudge factor' and I am not convinced it is needed. My suggestion is to not include cf, but instead estimate uncertainties in the various fits employed in the paper.

The factor cf is defined by fig 7 in which nINP from the CFDC set at a S of 105% vs the highest value of S which can be achieved before water droplets contaminate the signal. The idea is that not all aerosol that could serve as immersion INP do so at the lower S, but do so at the higher S. I do not think this is satisfactory. The upper limit to S is simply defined by the instrument limitation. If the instrument were redesigned to allow for a larger S again, would the INP concentration increase further, would cf then increase? It is well worth noting that the CFDC *may* undercount INP, but I do not think it should be used to correct INP concentrations.

I would like to see an error analysis. What is the uncertainty in the INP concentration predicted by the parameterisation based on the scatter of the data around the best fit lines. Looking at Fig 6, for example, there is significant scatter around the parameterization line – this probably accounts for more than a factor of 3 in uncertainty. Then when it comes to Fig 10 I suggest plotting the comparisons between measurement, the D10 scheme, direct field measurements and the prediction of the Niemand equation on 1:1 plots in which the uncertainties are indicated (probably for the dusty layer only; the point made about the INP in the MBL is valuable, but the key topic here is the mineral dust). I suspect that the cf=1 (i.e. no correction) curve would match the Niemand prediction within uncertainty in which case there is no need to introduce a correction factor. Similarly, the Niemand parameterization has some uncertainty with scatter of up to 1 order of magnitude either side of the best fit line (fig 3 of Neimand et al.); this should also be reflected in Fig 6. Given the uncertainty in both the

newdust parameterization and the Neimand line, I suspect there will be good agreement between the various data sets in Fig 10 without invoking a correction factor.

Authors' response [1]:

We agree that these are all important points to address, but we disagree with the reviewer on some key points that we have expended a lot of discussion on in this paper.

We first address the *cf* factor. The *cf* factor and the uncertainty of the parameterization are two distinct issues. Our use of correction, which we will prefer now to term calibration, is motivated by the following points:

1) Undercounting is clear and asymptotic toward higher RH_w on considering a host of experiments, not all of which can be shown. This behavior is demonstrated in this paper, but is also shown in Petters et al. (2009) for biomass burning aerosols. Hence, this is not a novel realization, but one we reinforce here, discuss in more detail some potential sources of this behavior, and strongly support as a factor that should be accounted for in any attempted parameterization of INP activation by immersion freezing for mineral dusts. Exact correction is not necessary, but a factor of 3 is inexplicable based on CFDC measurement uncertainties alone.

2) The application of a calibration factor for evaluating the maximum active fraction via immersion freezing using a CFDC-type instrument is supported not only by CFDC experiments in which RH_w is scanned, but through comparison to data obtained in surrogate cloud formation experiments in the AIDA expansion chamber.

3) We fail to see how attention to such detail undermines the validity of the measurement approach. We are obviously still learning in this field, and those lessons are important to share with a growing measurement community. We consider this to be a reasonable approach for estimating immersion freezing nucleation activity, but will now recommend calibration procedures for all future immersion-freezing related studies using the CFDC or any similar device. We will retain the *cf* factor as separate, rather than folding it into the actual parameterization, acknowledging this as a calibration factor that may vary in dependence on particular instrument and source aerosol type. This remains an area of important investigation for defining the meaning of measurements by any continuous flow INP instrument, as we have already concluded.

In defining the *cf* value, we have attempted to be transparent about factors that could lead to undercounting by CFDC type instruments, echoing and expanding on statements that have been made in previous papers. One may call it a fudge factor, but we wish to avoid that impression, since it would ignore our contention that there are plausible physical reasons for its existence. The existence of an artificial RH_w dependence of ice formation has been discussed previously in our publications, but it deserves special recognition so that it is clearly understood that that the sensitivity of INP number concentrations to RH_w is not likely to be readily resolvable as in a CCN instrument. Although research remains to fully elucidate the reasons necessitating an unrealistically high RH_w for the full expression of immersion freezing in a CFDC, we feel that parameterizations should use the calibration-corrected results because modeling studies seldom consider the outer limit of uncertainty when implementing parameterizations. RH_w dependence beyond a few percent is most certainly artificial, but account for this fact is needed to provide a parameterization that simply expresses the maximum immersion freezing activity following full CCN activation. It is a suggestion to err on the side of not undercounting. To meet the reviewer's concern, we further qualify our reasoning in applying correction/calibration entailed in *cf*.

We now use the term calibration in a number of places reference to *cf*. In discussing Fig. 2 within Section 2.1, we add:

This result suggests that unresolved factors are limiting the full expression/observation of immersion freezing nucleation in the CFDC until relatively high water supersaturation.

This point, in addition to adding proof that CCN activation is achieved at higher supersaturation (see below regarding a figure added to Appendix B in response to the comments of the second reviewer), emphasizes that there are limitations to full expression of immersion freezing that require calibration correction if one seeks to derive the maximum immersion freezing concentration (after all particles are within droplets), as is our goal.

We try to further emphasize this point in Section 2.3, where we have modified a statement to say,

The cf factor was not included in Tobo et al. (2013), by default being set to 1. The other equation coefficients could encapsulate this constant, but we will use it as a means to segregate instrumental calibration factors when assessing maximum immersion freezing concentrations or active fractions of mineral dust particles, as will be further addressed in this paper.

The reviewer is correct that the "upper limit to S is simply defined by the instrument limitation," albeit perhaps not a simple factor as it may depend on the particular design of this type of instrument. If the CSU CFDC instrument used in this study were redesigned to allow for separation of ice versus liquid particle signatures (e.g., lengthening the evaporation section of the column, or design of a suitable optical detector for small amounts of ice in a field of water droplets that would alleviate the need for the evaporation section), it is our expectation that INP concentration would not increase much further. This conclusion is stated on the basis of experiments where the highest RH_w was achieved and in other papers where particle size selection has allowed achieving even higher values (e.g., Petters et al., 2009). This is not an entirely objective answer to the question posed. However, a second piece of evidence already included, but neglected by the reviewer, is the AIDA expansion experiment data. The expansion-formed cloud provides the closest thing to ground-truth available (see response to review 2), with supersaturation created naturally for particle freely-suspended in the chamber. It is our strong belief that it is not fortuitous that this inferred "RH-delayed" activation of immersion freezing in the CFDC is also consistent with comparisons made with INP concentrations measured in AIDA expansion experiments. Finally, it is probably worth noting here that we have added a figure in response to a comment by reviewer 2 in order to demonstrate that full CCN activation of particles is likely achieved before the point of maximum supersaturation. This figure (Appendix B, Fig. B2) includes two additional experiments for which RH_w was ramped to higher values, well beyond the point of initial droplet breakthrough.

In the revised manuscript, the last sentence of Section 2.1 now states, *"Additional experimental support for the fact that nearly complete CCN activation and growth of mineral dust particles occurs in the CSU CFDC at RH_w between 105 and about 110% RH_w is given in Fig. B1."*

In Appendix B, as part of the discussion of the high RH_w offset for the maximal expression of immersion freezing, we write: *"Despite these concerns, evidence clearly exists for high CCN active fractions ultimately occurring in the CSU CFDC instrument for RH_w values close to the values used to define the maximum immersion freezing INP concentrations in these studies. Figure B1 shows two additional experiments from the ICIS-2007 studies where RH_w was raised to higher values to examine full droplet breakthrough, indicating CCN fractions up to 0.8. Similar freezing curves occurring for homogeneous freezing of solution droplets have*

also been previously demonstrated for the CSU CFDC instruments (DeMott et al., 2009; Koehler et al., 2009; Richardson, 2009; Richardson et al., 2010), indicating no special limitation on freezing high fractions of particles in these instruments. These results support the validity of the assumption that immersion freezing activity is assessed with the CFDC instrument in the present study."

The suggestion to add error analyses is an excellent one. We now state some statistical measures associated with the parameterization, add error bars on data and parameterization curves in all figures, and include the actual linearized parameterization results to show confidence intervals. As stated above, we in no way see the *cf* factor itself, representing an average calibration correction, as linked to these statistical uncertainties in the parametric fit. Parameterizations applied in models seldom consider the confidence intervals of the formulations applied. We provide them now, for those who might use the parameterization.

Hence, in Section 3.1, we now add *"Uncertainties represented by error bars on data points are twice the sampling error assuming Poisson arrival statistics for CFDC INP counts."* We have added a new figure (new Fig. 6) to graphically show the uncertainty of the predicted values. We have added error bars accordingly to Fig. 5 and to original Fig. 10 (now Fig. 11). These figures and the new captions are as shown below. In addition, we reorganize the discussion in Section 3.1 and write,

Comparison of predicted versus observed INP number concentrations for the entire data set are shown in Fig. 6. The r2 of the fit is 0.94, and the corresponding standard errors (a factor of ~2) and 95% confidence intervals (a factor or ~4) are also shown. Representative standard errors at specific temperatures are also mapped onto the predicted lines in Fig. 5.

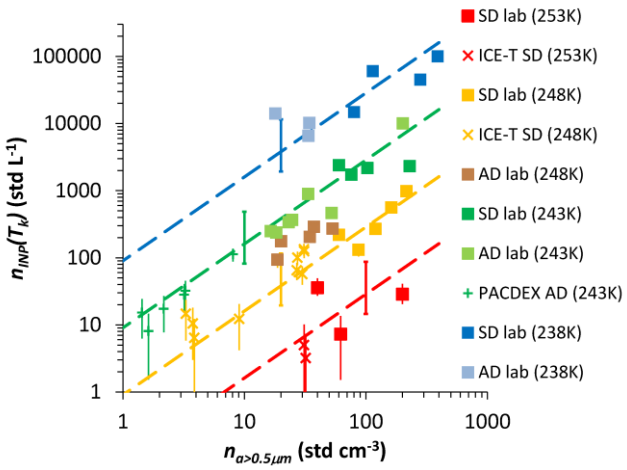

Figure 5. Relations between CFDC INP number concentrations measured at a nominal value of 105% RH_w and $n_{a>0.5\ \mu m}$ in laboratory (lab) and field (PACDEX and ICE-T) measurements of Asian (AD) and Saharan (SD) dust particles at temperatures of approximately 253, 248, 243 and 238 degrees Kelvin. Dashed lines are not best fits for each temperature, but are instead determined from the empirical fit given by Eq. (2) (cf = 1, α = 0, β = 1.25, γ = 0.46, and δ = -11.6). Uncertainties in observational data, given as twice the Poisson sampling error for the time-integrated samples, are shown by vertical error bars on data points. Note that at higher n_{INP} these error bars are not visible beyond the plotted point size. Representative measures of standard error in the predicted lines (see Fig. 6) are shown by capped error bars.

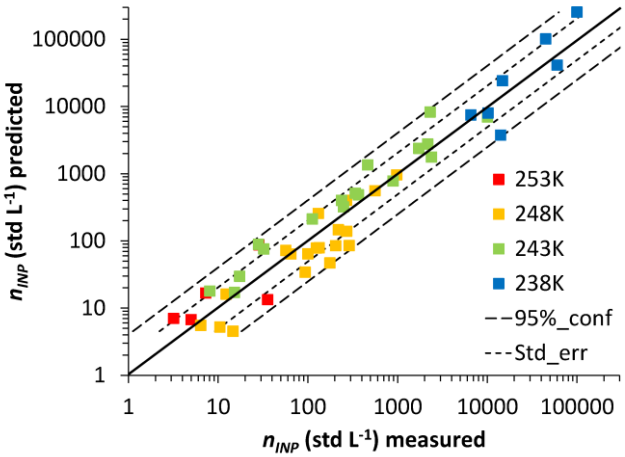

Figure 6. Prediction of Eq. 2 (cf = 1, α = 0, β = 1.25, γ = 0.46, and δ = -11.6), plotted versus raw field and laboratory data collected at 105% RH_w (Fig. 5), with lines added around the 1:1 line (solid) to indicate standard error (short-dashed) and 95% confidence intervals (long-dashed).

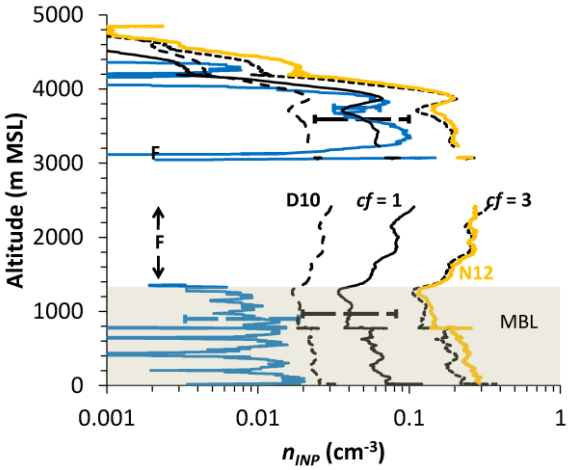

Figure 11. Comparison of ice nucleation data and parameterizations for data collected onboard the NSF/NCAR C-130 aircraft during the ICE-T study descent sounding through a Saharan dust layer shown in Fig. 4. CFDC INP data plotted as a 30-s running average at ambient conditions are given by the blue trace, the D10 parameterization is the long-dash trace, the solid black trace labeled cf = 1 is the uncalibrated parameterization derived using Eq. 2 (α = 0, β = 1.25, γ = 0.46, and δ = -11.6), and the short-dash trace labeled cf = 3 is the calibration-corrected parameterization with the same coefficients, both also corrected from STP to ambient INP concentrations. Uncertainties representing twice the Poisson sampling error of the 30-s running average data are given at two altitudes, and the standard errors of the cf = 3 prediction are shown at two nearby altitudes. Plotted for comparison is the parameterization of Niemand et al. (2012), using aerosol surface area and CFDC processing temperature as input. CFDC processing temperature cooled from 248 K at 5 km to 246 K at landing, while CFDC calculated RH_w at the lamina position was maintained at 105±0.5%. The shaded region represents the marine boundary layer (MBL). Label F indicates that CFDC sample air was being filtered. The data gap is when CFDC flow was shut off to remove an ice crystal impactor.

We do not agree with the suggestion to revise Fig. 10 (now Fig. 11). We feel that the representation as a vertical profile is much more powerful and intuitive.

Reviewer's comment [2]:

Concerning the point made about 'mineral dust particles from locations as separate as the Saharan or Asian regions may be parameterized as a common particle type for numerical modelling purposes'. This is an interesting observation and as the authors point out in line with what has been suggested previously by Neimand. A brief discussion of why this is the case is needed. An explanation is that there is a common component of these dusts which triggers ice formation. Atkinson et al. (Nature, 498, 2013, doi: 10.1038/nature12278) suggest that this minor component is feldspar which is ubiquitous in natural soil dusts.

Authors' response [2]:

We add the requested discussion of the matter that certain dust components such as K-feldspar could be playing a role in controlling the relative uniformity of mineral dust activation properties globally. It is less clear how such chemical/mineralogical differences could be manifested in a relation that uses only temperature and aerosol concentrations above 0.5 μm as the controlling quantities. Hence, we have added to the discussion within the conclusions to say,

"The reason for this result is not entirely clear, given the clear mineralogical differences present in and transported from different desert regions (Murray et al., 2012). Possibly, the relatively high abundance (>20% by mass) of more highly ice-active specific components of dusts, such as feldspars, from both Asian and Saharan regions (Atkinson et al., 2013) drives this result. Nevertheless, it remains to be seen that this conclusion is fully consistent with the unifying role of aerosol concentrations at >0.5 □m or total surface area of mineral dust particles on determining INP number concentrations, since many other mineral components make up the balance of dust particle mass. It remains for additional measurements at different locales to further evaluate this conclusion regarding the relative uniformity of INP properties of mineral dust particles globally or, alternately, to demonstrate the special utility of mineralogical-specific parameterizations."

Reviewer's comment [3]:

Keys within figure 5 and 7 would be helpful for the reader. Having to refer to the caption takes longer than referring to a key.

Authors' response [3]:

The new figures (5 and 8) have been revised accordingly, to make them easier to decipher.

Authors' responses to comments by Referee #2

We thank Anonymous Referee #2 very much for his/her comments. Below are our point-by-point responses.

Reviewer's major comment [1]:

The developed parameterisation is not able to quantify immersion freezing ice nucleation activity in a microphysical manner. Eqs. (1) and (2) provide empirical relationships between the INP concentration detected with the CSU-CFDC, temperature and particle concentration larger 0.5 μm. These relationships might be limited to their experimental conditions, but not generally valid. This limitation should be reflected in the paper.

Authors' response:

This is a good point. We do state upfront, starting with the Abstract, that developing an empirical parameterization is a part of the study. We discuss reasons for and the potential utility of this approach in the Introduction. However, to make the nature of the parameterization clear, and to frame the specific limits of its use, we change the words "simple parametric" in the abstract to say "empirical", and we now state in the paper the specific temperature range over which data were used. Additionally, we add statement that extrapolation outside of this regime cannot be expected to be reliable.

Thus, at the end of Section 3.1 we write*: "The parameterization developed herein is strictly valid where data were available, between 238 and 252 K, and use to warmer temperatures represents pure extrapolation."*

At the end of Section 3 we write: *"While providing confidence that both parameterizations can thus be used to describe atmospheric ice nucleation by mineral dust particles specifically in the temperature ranges for which they were developed, we note that comparison to ice formation in atmospheric clouds has yet to be examined"*.

In the first paragraph of the Conclusions section we write: *"Use of the parameterization to warmer temperatures necessarily entails extrapolation of the present results."*

Reviewer's major comment [2]:

An experimental demonstration that CFDC experiments running above water saturation or 105% RH_w respectively, approximates immersion freezing is missing. Mainly the unknown CCN active particle fraction is an issue. E.g. Welti et al., 2014 observed only 4% CCN active particle in their CFDC experiments covering a wide temperature range and RH_w up to 110%.

Authors' response:

We add more information and a new figure to the paper in response to this point, as discussed further below. We could not find information in Welti et al. (2014) to corroborate the last statement of the reviewer. Welti et al. (2014) show details of CCN active fractions of no more than 0.04 at 100% RH_w in their Fig. 6, and they then additionally interpret the ice nucleation signals above water saturation as representing other processes such as deposition and condensation freezing nucleation. No further attempt is made to discern or interpret ice nucleation at higher water supersaturations as immersion freezing, with the immersion freezing contribution being entirely defined by results from separate/previous experiments with a different experimental device. Low CCN activation at 100% RH_w is described to result from CCN activation delays in the case of mineral dusts at lower temperatures, at least ones that do not have associated soluble material. Nevertheless, they cautiously state, "An open question that remains is to what degree and at what saturation mainly insoluble dust particles will act as CCN at low temperatures." We take a different approach. While deposition and condensation freezing processes may be possible in the water supersaturated regime, in addition to immersion freezing, no direct resolution of the existence of multiple processes in single experiments has yet been demonstrated. Welti et al. (2014) compare different experiments (ZINC versus IMCA-ZINC) to derive the apparent non-immersion freezing contribution. This mimics such subtraction and calculation methods used in earlier studies that they reference. We assume that full CCN activation and growth must occur once an elevated RH_w is reached in the CFDC. We already point out that the expression and detection of CCN activation and freezing response in a CFDC-type instrument could depend on factors that are specific to a given device. It is our expectation that a CFDC should produce CCN activation in the same manner as a CCN instrument does, albeit with limitations imposed by delivery of the aerosol lamina, the variable supersaturation profile of a flow diffusion chamber, and other chemical kinetic factors. As evidence that CCN activation ensues well ahead of 110% RH_w and prior to the point that we infer maximum ice activation in the CSU

CFDC (i.e., it is occurring already at 105%, but the full expression is not seen until higher RH_w), we now provide references and a new figure (Fig. B1) shown below. We also note that previous studies suggest no impedance of water uptake and homogeneous freezing in the CFDC, when the instrument is operated to provide sufficient growth times at low temperatures (DeMott et al., 2009; Koehler et al., 2009; Richardson, 2009; Richardson et al., 2010). Those studies already suggest up to 70% of aerosols of varied hygroscopic properties achieving appropriate water uptake to freeze as nearly dilute cloud droplets at -40°C, or as more concentrated solution droplets at lower temperatures. The Koehler et al. study is notable for demonstrating the varied RH_w for which full freezing ensues in dependence on the water uptake properties of particles. We add here results of two more RH_w scans from the ICIS-2007 studies conducted for desert dust particles at a slightly warmer temperature for which there was only very little freezing observed and for which RH_w was raised to much higher values. These experiments were not otherwise used for the parameterization development in the paper. The new figure and caption are here:

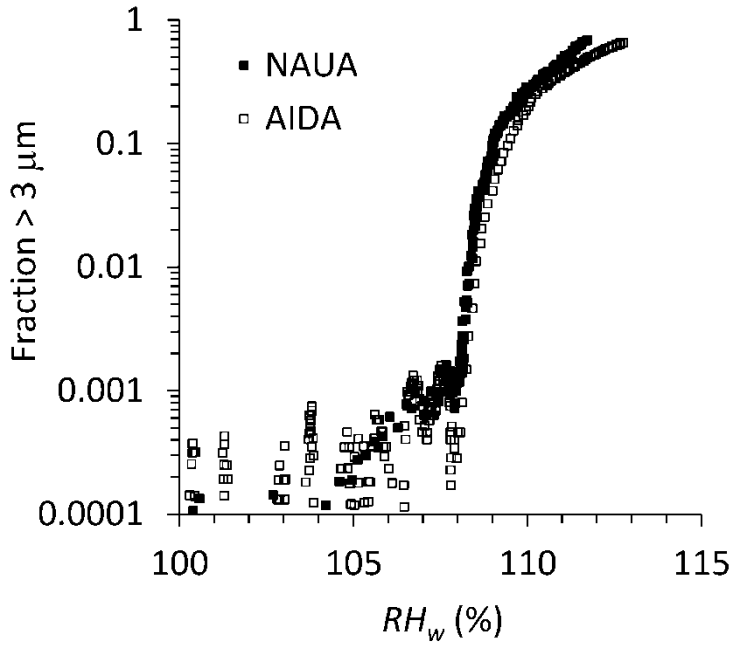

Figure B1. As in Fig. 2, raw 1 Hz CFDC data from an ICIS-2007 experiment on the fraction of total aerosol concentrations (measured by a CPC) appearing at OPC sizes above 3 µm during RH_w scanning for two experiments at -21°C when processing particles from a dust sample that had been collected following a dust storm in Israel (Kanji et al., 2011). The data termed NAUA was sampled following dispersion into a 4 m^3 aerosol chamber, with concentrations of approximately 5000 cm^{-3} present at the time of sampling. The data termed

AIDA was sampled directly from the AIDA expansion chamber prior to a cloud expansion, when the total particle concentrations were approximately 100 cm^{-3}. Water droplet breakthrough of the CFDC evaporation region occurs at ~108% in each case and progressively more activated cloud droplets survive through the evaporation region as RH_w is increased further.

Elevation of RH_w well beyond the point of water droplet breakthrough at ~108% RH_w in Fig. B1 shows that the fractions persisting at water droplet sizes following the evaporation section of the CFDC increase up to 0.8 at higher supersaturations. Absent removal of the evaporation section (i.e., maintenance of supersaturation to the bottom of the instrument) to demonstrate the existence of water droplets in the growth section of the CFDC, not possible in this particular study, these data strongly support that unimpeded immersion freezing on fully activated water droplets should be observable in the CFDC at a point prior to the breakthrough RH_w. For this reason, we believe that our interpretation of experimental results is plausible, with the Welti et al. study providing a contrasting viewpoint.

In the revised manuscript, the last sentence of Section 2.1 now states, *"Additional experimental support for the fact that nearly complete CCN activation and growth of mineral dust particles occurs in the CSU CFDC at RH_w between 105 and about 110% RH_w is given in Appendix B, Fig. B2. We may contrast our assumptions and approach in this regard to that of Welti et al. (2014), who used separate experiments to define immersion freezing fractions and then applied calculation and subtraction methods to interpret and attribute additional INP fractions freezing as contributions from a condensation freezing process at RHw > 100%."*

In Appendix B, as part of the discussion of the high RH_w offset for complete immersion, we write: *"Despite these concerns, evidence clearly exists for high CCN active fractions ultimately occurring in the CSU CFDC instrument for RH_w values close to the values used to define the maximum immersion freezing INP concentrations in these studies. Figure B1 shows two additional experiments from the ICIS-2007 studies where RH_w was raised to higher values to examine full droplet breakthrough, indicating CCN fractions up to 0.8. Similar freezing curves occurring for homogeneous freezing of solution droplets have also been previously demonstrated for the CSU CFDC instruments (DeMott et al., 2009; Koehler et al., 2009; Richardson, 2009; Richardson et al., 2010), indicating no special limitation on freezing high fractions of particles in these instruments. These results support the validity of the*

assumption that immersion freezing activity is assessed with the CFDC instrument in the present study."

Reviewer's major comment [3]:

Comparing to the AIDA expansion chamber with an uncertainty in the CCN active particle fraction of +/-30% (Niemand et al., 2012) seems not optimal. A comparison to immersion freezing experiments ensuring particles are immersed in droplets (IMCA, coldstage, EDB, DSC) would be interesting.

Authors' response:

Nearly every method to induce and observe immersion freezing entails some non-ideality or experimental issues. We do not possess an IMCA, and we promote in this study that we do not necessarily require one to effectively observe immersion freezing. Furthermore, we consider that the most straightforward natural simulation of immersion freezing must be for a volume of air undergoing expansion that is not in close contact with walls, or subjected to strong thermal gradients, electric fields and so forth. Hence, a larger expansion chamber like AIDA has long been envisioned as a standard for ice nucleation studies. We do not consider the AIDA uncertainty in droplet active fraction of +/-30% at low aerosol concentrations to be particularly limiting, and during restricted periods as shown in our very typical example of experiments used in this study (Fig. 3), variability is often less than that. Description of experiments comparing the CFDC instrument to a classical immersion freezing device for other various dusts are in preparation for separate publication at this time.

In Section 2.2, we amend the discussion as: *"Full activation of aerosol into cloud droplets is achieved in AIDA (±30% maximum deviation, as noted by Niemand et al., 2012) and ice active fraction…"*

Reviewer's major comment [4]:

Another option would be a characterisation following the homogeneous freezing curve to determine the fraction of CCN active particles. From the discussion in Appendix B it appears

that this is not feasible due to the lower temperature in the evaporation region. Is that the case?

Authors' response:

The reviewer interprets correctly that the CFDC instrument used in this study contains an evaporation section in which the temperature of the air decreases as droplets are evaporated away. This is not the most ideal for examining the homogeneous freezing response of an aerosol population, but since the RH_w decreases at the same time as temperature in this region, there may be little impact. This type of CFDC and a version of the CSU CFDC design for which the wall temperatures were not altered to induce evaporation in the lower portion of the instrument have been used in studies of homogeneous freezing (DeMott et al., 2009; Koehler et al., 2009; Richardson, 2009; Richardson et al., 2010), and these show the ability to detect homogeneous freezing of CCN nearly ideally. We add this information with additional information on CCN activation (next comment) into the revised manuscript.

Repeated our response above, in Appendix B, where much of this discussion already appears, we write: *"Similar freezing curves occurring for homogeneous freezing of solution droplets have also been previously demonstrated for the CSU CFDC instruments (DeMott et al., 2009; Koehler et al., 2009; Richardson, 2009; Richardson et al., 2010), indicating no special limitation on freezing high fractions of particles in these instruments. These results support the validity of the assumption that immersion freezing activity is assessed with the CFDC instrument in the present study."*

Reviewer's major comment [5]:

Has the active fraction in the breakthrough regime been observed to reach 100%?

Authors' response:

As noted already, we have confirmed active fractions approaching 100%, although usually up to 20% less. Full confirmation of such activation at a specific RH_w above 100% would require use of a detection method that could differentiate liquid and ice particles. This has not yet been achieved for our instrument, and therefore we now qualify that full CCN activation is by the inference of other experimental evidence, such as the RH_w scans shown in the new figure.

Reviewer's major comment [6]:

According on Fig.2 it seems that only 4% of the particles are CCN active. Is that the case?

Authors' response:

No, this is not the case based on our experience in performing many RH_w scans with the CSU CFDC, including the additional ones now shown from the same experimental period as some of the laboratory studies used for analyses in this paper. The case shown is Fig. 2 was terminated prior to realizing full water droplet breakthrough of the evaporation section, as full definition of that breakthrough signal was not sought in most experiments. Rather, it was simply recognized and then the experiments were terminated. We used the case in Fig. 2 as the experimental example due to its use in the study as one of the Saharan dust experiments. As a future recommendation, based on both reviewer comments, we now amend statements in our Conclusions as, *"Nevertheless, these results have implications for the design and operation of any CFDC-type ice nucleation instrument, suggesting careful characterization of ice nucleation response to RHw for any particular device and different INP types that compose natural populations. In particular, scanning up to and beyond the RH_w for droplet breakthrough to establish CCN activation (e.g., Fig. B1) is recommended. This will more clearly define the upper RH_w limit for assessing ice active fraction versus temperature uniformly for any INP type being tested and CFDC instrument type being used.*"

Reviewer's detailed comment [1]:

17368, line 19 and Fig. 3: What is the RH_w for CCN activity setting in AIDA and how long does it take to form a cloud once this RH_w is reached?

Authors' response:

As in the atmosphere, there are no highly accurate measures of RH_w inside a cloudy volume of air. With a water vapor concentration accuracy of 3-5% using the AIDA tunable diode laser hygrometer (Fahey et al., 2014), the CCN activation RH_w cannot be accurately measured or validated in AIDA. The matter of most relevance is what proportion of particles are put into liquid drops at the point of cloud activation that are then further cooled. We suggest that this proportion is essentially 1, with the stated 30% uncertainty. Based on Fig. 3, which is a common example for the present studies, full activation, as measured by cloud particle counters, is achieved in less than 15 s. No special addition to the text is made, except to note that in the figure caption for Fig. 3 that the data points are at 5 s intervals.

Reviewer's detailed comment [2]:

17370: One particular feature of Eq. (2) is that it predicts the fraction of particles (which are larger than 0.5μm) initiating ice formation to increase with increasing n_a>0.5μm at a given temperature. Accordingly fractions above 1 can result for high n_a>0.5μm and low temperatures. Does this indicate that the parameterisation is only valid for a specific range of particle concentration or activation of smaller particles? Where does the choice of a>0.5μm originate from?

Authors' response:

One must be careful not to confuse a parameterization that simply relates INP number concentration to the number concentration of particles of diameter > 0.5 μm with the population of aerosols of all sizes that are available to serve as INP. Hence, active fractions > 1 with reference to 0.5 μm particles are allowed, the occurrence of which is fully consistent with the fact that 0.5 μm is the observed mode size of for natural INP over the course of a number of studies. Smaller INP do exist. The use of this INP reference size, motivated by the observed mode size of natural INP and natural dust particles, and historic studies noting the relation between INP concentration and the concentration of all aerosol particles exceeding a certain size, is discussed in detail in DeMott et al. (2010). Practically, active fractions exceeding 1 in comparison to n_a > 0.5 μm are restricted to very low temperatures, as it is the case that INP concentrations equivalent to only a few to ~10 percent of n_a > 0.5 μm are typically active for natural dust particles at -30°C. However, the parameterization shown in this paper should be restricted for use in the mixed-phase cloud regime at temperatures warmer than homogeneous freezing occurs. We try to clarify this point now.

"The parameterization developed herein is strictly valid where data were available, between 238 and 252 K, and use to warmer temperatures represents pure extrapolation."

Reviewer's detailed comment [3]:

17382 line 13-15: Looking at Fig. 1 the evaporation region is at -35C for an experiment at -30C and 105% RH_w. But the droplets seem to only be cooled down by 3 and not by 5 C, why?

Authors' response:

This is due to the kinetics of heat and mass transfer. Given more time in the evaporation region, full cooling would be achieved. Temperature in the aerosol lamina relaxes in time toward the cold wall temperature.

Reviewer's detailed comment [4]:

17383 line 15-17: The statement that the RH_w dependence can be ignored should be supported by more discussion. Fig.2 shows that not all particles are activated at 105% and that the activated fraction is increasing with increasing RH_w. These results show a RH dependence and thus contradict the above statement. Doesn't Fig.2 also disprove the Petters et al (2009) assumption on p. 17367/17368 that the nucleation mode observed at 105% RH_w is immersion freezing? It seems necessary to reconcile these apparently contradicting results.

Authors' response:

What appears needed is for us to more clearly articulate this discussion. The RH_w dependence shown in Fig. 2 is the real expression seen in raw CFDC data. However, it's source for physical reasons that relate entirely to the instrument and particle response within it is the reason for ignoring specific attirubution of INP concentrations to specific RH_w values during the asymptotic increase of INP concentration with RH_w, and in attributing full immersion freezing activity instead to the maximum value achieved prior to water droplet breakthrough. Petters et al. (2009) promote the use this high RH_w value as reference, not 105% RH_w. The 105% value is a value that is almost always practically achievable in any ambient sampling scenario with a CFDC. For example, one would not want to flirt with entering the droplet breakthrough regime if they are collecting ice crystals from activated INP onto an impactor for inspection of INP composition. That would contaminate the sample with droplet residues. Furthermore, the breakthrough RH_w values depend somewhat on temperature (Richardson, 2009). Hence, these higher RH_w values are simply avoided in practical sampling, although this might motivate one to alter the CFDC design in the future for access to RH_w values that are not usually deemed of relevance (yet they are, at least for quantifying immerion freezing – that is the point). In a laboratory setting we are able to assess the full activation behavior with experimental RH_w up to the point of drop breakthrough, and we use that information here to gain better quantification of immersion freezing of mineral dust particles. We are literally calibrating the values obtained at 105% for these types of INP and we are saying that there is a correction necessary.

For this reason, we now emphasize *"calibration"* instead of *"correction"* in relating immersion freezing observed at 105% to that at an RH_w deemed to represent full CCN activation. In the abstract we modify statements to read, "Measurements made with the Colorado State University (CSU) continuous flow diffusion chamber (CFDC) when processing mineral dust aerosols at a nominal 105% relative humidity with respect to water (RH_w) *are taken as a measure of the immersion freezing nucleation activity of particles.*" In Section 2.1, prior to referencing more detailed discussion of Appendix B on why we believe that freezing at 105% is not the full expression of immersion freezing, we modify a statement to read, "Finally, a reference condition was placed on the processing RH_w *deemed representative of immersion freezing nucleation.*" And in discussing Fig. 2, we now write, *"This result suggests that unresolved factors are limiting the full expression/observation of immersion freezing nucleation in the CFDC until relatively high water supersaturation."* Changes related to CCN activation in the CFDC have already been mentioned above.

Reviewer's detailed comment [5]:

Fig.2: As pointed out before the active fraction is increasing with increasing RH_w. This is either a disagreement to an immersion mode mechanism taking place or an indication of incomplete CCN activation of the particle population.

Authors' response:

We believe that we have described evidence that this ensues due to requiring RH_w in excess of that presumed necessary for full CCN activation of the particle population, kinetics of droplet growth, non-uniformity of exposure of all particles to exactly the same RH_w, and so forth. It is difficult to know how to further emphasize this point.

Reviewer's detailed comment [6]: Fig.3: Why does the scale of the number fraction go up to 3? At what RHw is complete CCN activation at -30 C detected in AIDA? Please consider adding a third panel showing the RHw, RHi conditions prevailing during the experiment.

Authors' response:

The scale goes to 3 so that the maximum values around 1 are clearly visible. This shows that CCN activation is nearly complete in the volume of the AIDA cloud. Plots including RH_w are already shown in Niemand et al. (2012) and in other papers from the AIDA group, so we choose not to add a panel with that measurement here. What we consider most relevant are the facts that full CCN activation occurs, followed by ice formation as the parcel cools. If the reviewer is seeking to somehow point out differences in the ice nucleation signal response in the AIDA expansion chamber versus the CFDC, we can only note that one should not expect to see the exact same behavior in simulated cloud parcels as seen in a CFDC. See our earlier responses on this topic, and the extended discussion of the RH_w response in the manuscript.

Reviewer's detailed comment [7]:

Fig.5: A red line for high- blue for low temperature would be more intuitive. Some

dashed lines are not reproduced. Please also add error bars (in both directions).

Authors' response:

We have altered the color scheme as requested. Dashed lines have been improved in the new figure version. Error bars are now added to data points. Representative error bars, representing standard prediction error are added now on the parameterization lines (see response to Reviewer 1).

Reviewer's detailed comment [8]:

Fig. 6: I would expect D10 to be a straight line in this figure. Why isn't it, i.e. where does the scatter come from? Please add in the caption that $cf = 1$ was used.

Authors' response:

Thanks for this comment. D10 points vary slightly from a straight line due to use of the actual temperatures of observations, which are allowed to vary slightly (within 1°C) from the reference processing temperatures used for comparison, as stated in the methods discussion. We note this now and add the $cf = 1$ qualifier in the figure caption. This figure is now Figure 7.

Reviewer's detailed comment [9]:

Fig. 7: RH_{max} is instrument specific and therefore an arbitrary condition to compare to. The active fraction would increase further for higher RH_w as can be seen e.g. in Welti et al, 2009. Isn't the correction factor an indication that the mechanism looked at is different from immersion freezing?

Authors' response:

RH_{max} is not arbitrary if justified within a certain range for a specific CFDC type instrument. For the CSU CFDC, further increase may occur, but we contend that we are reaching a plateau by 109% RH_w. This is consistent with the additional evidence and figure we now provide (see response to major comment #2) that shows that a high proportion of particles are clearly within droplets before this high value is achieved in the CSU CFDC. The reviewer is correct that this is a function of the particular instrument, and a matter for each investigator to explore and quantify. Given evidence that CCN activation is achieved at values below 109% RH_w in the CSU CFDC for the dust particles sampled, we cannot justify that we are observing a different mechanism of ice formation than immersion freezing at that point. On the otherPlease note that Fig. 7 will now be Fig. 8.

Reviewer's detailed comment [10]:

Fig. 10: Should the RH_w range be 105 +/-3% as stated in the text?

Authors' response:

We will clarify in the figure caption that the calculated lamina RH_w was held within the stated range of 0.5%. This simply says that good control on the target RH_w was maintained. We now provide more accurate discussion of the RH_w uncertainty in the methods section of the paper on the basis of temperature-dependent information provided in Richardson (2009).

We state in Section 2.1, *"*An additional consideration in selecting this value *is that RH_w uncertainty, as estimated and extrapolated from Richardson (2009), is ±1.6, 2 and 2.4 % at -20, -25, and -30°C, respectively (Hiranuma et al., 2014)."*

Reviewer's detailed comment [11]:

Fig. A1: b) The scale "Fraction of particles observed" should end at 1.

Authors' response:

We prefer a scale which allows resolution of the values reaching their full value of 1.

References:

Fahey, D. W. et al.: The AquaVIT-1 intercomparison of atmospheric water vapor measurement techniques, Atmos. Meas. Tech., 7, 3177–3213, 2014.

Hiranuma, N. et al.: Supplement of: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of seventeen ice nucleation measurement techniques, Supplement of Atmos. Chem. Phys. Discuss., 14, 22045–22116, doi:10.5194/acpd-14-22045-2014-supplement, 2014.

Koehler, K. A., DeMott, P. J., Kreidenweis, S. M., Popovicheva, O. B., Petters, M. D., Carrico, C., Kireeva, E., Khokhlova, T., and Shonija, N: Cloud condensation nuclei and ice nucleation activity of hydrophobic and hydrophilic soot particles, Phys. Chem. Chem. Phys., 11, 7906 – 7920, 2009.

Richardson, M. S., 2009: Making Real Time Measurements of Ice Nuclei Concentrations at Upper Tropospheric Temperatures: Extending the Capabilities of the Continuous Flow Diffusion Chamber, Ph.D Dissertation, Colorado State University, 262 pp.

Richardson, M. S., DeMott, P. J., Kreidenweis, S. M., Petters, M. D., and Carrico, C. M., 2010: Observations of ice nucleation by ambient aerosol in the homogeneous freezing regime. Geophys. Res. Lett., 37, L04806, doi:10.1029/2009GL041912.