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.

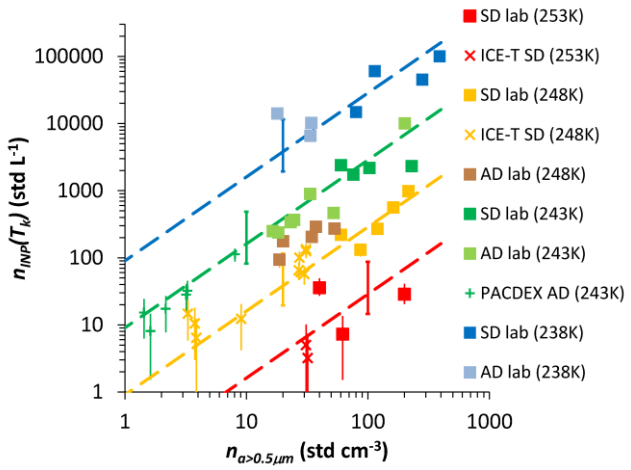

Figure 5. Relations between CFDC INP number concentrations measured at a nominal value of 105% RH_w and $n_{a>0.5\ \mu m}$ in laboratory (lab) and field (PACDEX and ICE-T) measurements of Asian (AD) and Saharan (SD) dust particles at temperatures of approximately 253, 248, 243 and 238 degrees Kelvin. 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.

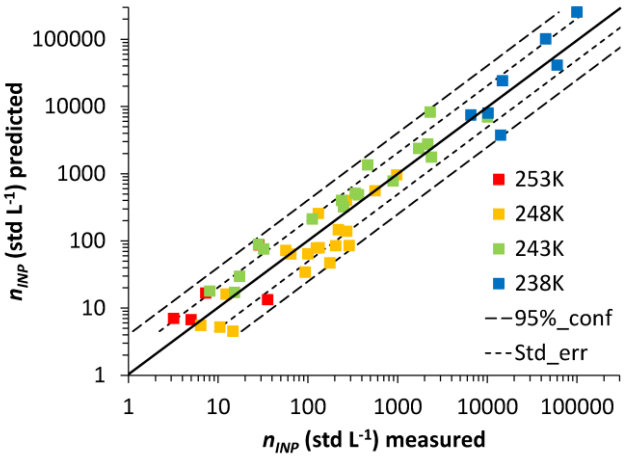

Figure 6. Prediction of Eq. 2 (cf = 1, α = 0, β = 1.25, γ = 0.46, and δ = -11.6), plotted versus raw field and laboratory data collected at 105% RH_w (Fig. 5), with lines added around the 1:1 line (solid) to indicate standard error (short-dashed) and 95% confidence intervals (long-dashed).

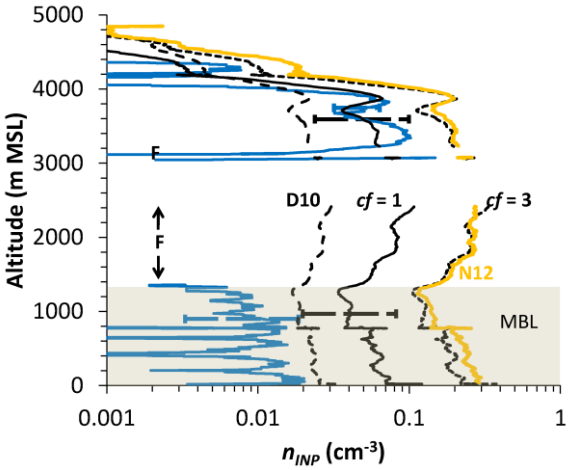

Figure 11. Comparison of ice nucleation data and parameterizations for data collected onboard the NSF/NCAR C-130 aircraft during the ICE-T study descent sounding through a Saharan dust layer shown in Fig. 4. CFDC INP data plotted as a 30-s running average at ambient conditions are given by the blue trace, 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.