We would like to thank Paul DeMott and the anonymous Reviewer for having read the revised paper. We sincerely appreciate the insightful second review by Paul DeMott. We found it very helpful to further improve the revised version of the manuscript. Point-by-point replies to the comments are below.

For clarity and easy visualization, the Referee's comments are shown from here on in black.

> The authors' replies are in blue font with an increased indent below each of the referee's statements.
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> > The relevant changes in the revised manuscript are below in green. If just a part was added to an existing sentence, then the added part is underlined. All line and page numbers in normal font refer to the first version of the revised manuscript. All line and page numbers **in bold** refer to the second version of the revised manuscript.

Replies to the second review by Paul DeMott

General Comments

I greatly appreciated the detailed responses to my comments, and the subsequent revisions. I have a few issues of concern still, primarily in further discussing the superposition of the D15 parameterization on plots using aerodynamic diameter as the basis, and the *cf* factor applied in using that parameterization. In combination, these two factors will, I believe, bring the parameterization nearly fully in line with the observations under SD conditions. This is as it should be, if the parameterization has any validity. One could imagine that it should fail in capturing all dust INPs because it does not fully capture those active larger than 2.5 microns(aerodynamic), but I have two comments about this conjecture. First, the D15 relationship was developed from both free tropospheric (elevated from the ground) and laboratory data for which there was a strong relation with >500nm particle concentrations under situations that were totally composed of, or strongly perturbed by, desert dust. In fact, I think that the present study finds the same thing! Note the equivalent correlation coefficient for your linear regressions in SD situations, for INPs versus 0.5 and 2.0 micron aerosol concentrations. This would not be the case if INP concentrations under SD situations were dominated by particles larger than 2.5 microns, would it? This brings me to the second point. The present studies say nothing about the actual size of INPs at the site used in this study. Some investigators have found larger INPs at surface sites, albeit likely because of influences of INPs that are not mineral dust, and partly because the measurements are focused in the near-surface boundary layer where larger particles are always found. Mineral dust is clearly not the major contributor at the site in this paper either, except under SD scenarios. What the current study does show is that in order to obtain a clean relation between aerosol concentrations and INP concentrations under most situations, one must reference a larger particle size for parameterization. That does not permit an assured conclusion that the INPs are always typically larger than 2 microns. You said it yourself that INPs are but a minute fraction of the total aerosol. It could simply mean that other factors enter to populate the smaller size ranges with particles that are not INPs and do not vary in-kind with them. This is another important distinction in my opinion. I feel that you confuse the

issue in deciding that larger INPs are the reason that you have to go out to 2 microns to get a good correlation.

We address all the general concerns raised above in their more specific versions below. Only the "conclusion that the INPs are always typically larger than 2 microns" is not taken up again as a specific comment. Therefore, we address it here.

We reformulated what we think is the contested sentence (i.e. "However, choosing the actual size range of $INPs_{-15}$ for the parametrisation can further improve the predictions.") in the Conclusion section as follows:

(P6L156; **P6L163**) However, relating [$INPs_{-15}$] to the number concentration of larger particles can further improve the predictions, which is not to say that $INPs_{-15}$ are always in such a size range.

Specific Comments

1) Regarding changes made about previous literature on the role of ice formed and growing at temperatures above about -15 °C, I have a suggestion. The meaning of the revised statement that "...although other temperatures would benefit from future investigations" is somewhat ambiguous. I think you could say that investigations would benefit from relation of measurements to overall cloud thermal structure, which may at times include lower cloud top temperatures.

We agree that this statement is somewhat ambiguous, and we like your suggestion. We rephrased this part of the sentence as you suggested:

(P2L32; **P2L32**) We, therefore, in this work, focus on INPs active at that temperature, although future studies would benefit from relating measurements to overall cloud thermal structures, which may at times include lower cloud top temperatures.

2) Related to making clear that D15 is strictly for application on mineral dust dominated populations, somewhere around the introduction of Equation 1 it needs to be stated that the equation will here be applied to all particles at sizes larger than 500 nm. I understand that the concentration parameter is explicitly defined in Equation 1, but I mean that it should be said in words that although the parameterization is strictly for mineral dusts, it will be applied to all particles. The reason for doing this is to note later that one only expects this parameterization to be valid as related to data under strong dust influences (e.g., SD here).

We specified in the text that Equation 1 will be applied to mineral dust influenced particles larger than 500 nm only. Furthermore, we now focus on comparing D15 with our SD data and make only a very cautious comparison to data not strongly influenced by mineral dust (see reply below the next comment).

(P3L82, **P3L83**) [$INPs_{-15}$] estimates based on D15 were calculated as:

$$INP_T = cf * {n_{0.5}}^{\beta} * e^{\gamma*(-T)+\delta}, \quad (1)$$

where β = 1.25 , γ = 0.46, δ = -11.6, T is the temperature in degree Celsius, INP_T the ice nucleation particle concentration (std L^{-1}) at T, and $n_{0.5}$ the

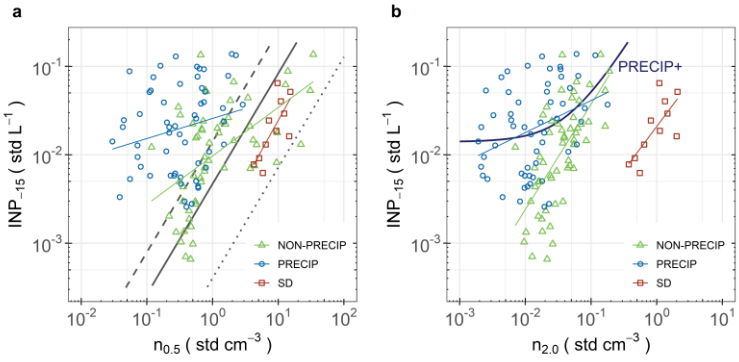
number concentration of aerosol predominantly consisting of mineral dust particles with a physical diameter > 0.5 µm (std cm^{-3}).

3) The justification for *cf* = 1 is incorrect, I believe. You say that no calibration factor is required "because INPs were observed in immersion mode (via a drop freezing assay) and not for instance, in a continuous flow diffusion chamber, where...only part of the INPs passing the instrument may become immersed in liquid droplets." This is exactly why *cf* = 3 is needed. If one were comparing data directly from a CFDC to the parameterization, then *cf* = 1 is what you would want to use, as was done in that paper in 2015. If one is using the parameterization in a model and applying it to the dust distribution, or if one is comparing to a method that captures all dust INPs active by immersion freezing, then *cf* = 3 is what one wants/needs to use. It is the full intention of the parameterization, based strongly in the results presented in that paper.

Thanks a lot for pointing that out. We indeed misunderstood the calibration factor choice in the last version of the manuscript. We rephrased this in the text and now show in Fig. 3a predictions based on three *cf* values: 3 and 1. In addition, we added a prediction based on a *cf* = 0.086, a value reported by Schrod et al. (2017) done in the Mediterranean region. We changed Fig. 3a accordingly. Furthermore, we corrected the unit of the label on the x-axis, which was erroneously the same as on the y-axis (std L^{-1}) to (std cm^{-3}).

(P3L85-L88; **P4L96**) The calibration factor *cf* accounts for so-called instrument-specific calibration and is suggested to be three (*cf* = 3) to predict maximum immersion mode atmospheric [INP] (DeMott et al., 2015). Schrod et al. (2017), who collected samples with an unmanned aircraft system in the Mediterranean region with substantial Saharan Desert dust influence, used it as a mathematical degree of freedom when fitting Eq. 1 to their observations.

(P4L120-P4L125; **P5L128**) In general, [$INPs_{-15}$] in non-precipitating and precipitating (not dominated mineral dust) air masses were higher than in mineral dust dominated air masses for the same [$n_{0.5}$] (Fig. 3a). The observed slope for SD air masses was the same as that predicted by the D15 parametrisation. The offset of the D15 curve depends on the calibration factor (*cf*, Eq. 1). Observed SD data were between the D15 curves with *cf* set to 1 and to 0.086, respectively. The latter value is reported in Schrod et al. (2017), who sampled the Saharan Dust Layer above Cyprus with a drone up to 2850 m a.s.l.



(Figure 3, **Figure 3**) […] The gray lines show the D15 parametrisation extrapolated to -15 °C and corrected for the difference between physical and aerodynamic diameters (see Methods section) with three different calibration factors (Eq. 1): *cf* = 3 (dashed), cf = 1 (continuous), and *cf* = 0.086 (dotted). The latter value was the best fit found by Schrod et al. (2017) for observations of Saharan dust above Cyprus.

4) Regarding the explanation of the upper bound of concentrations, was dilution not possible, or simply determined not to be desirable? Perhaps say that dilution was not used in order to extend the upper range.

Dilutions were not considered to be reliable because of unavoidable storage and its effects. The sampling campaign was at times extremely busy and our target was to analyse the (undiluted) samples as quickly as possible after sampling. Therefore, dilutions had to wait.

5) Regarding the use of aerodynamic diameter because there is no way to know shape factors and density, I want to stress again that for the purpose of showing the D15 prediction, this paper should be focused on comparison to mineral dust dominated cases. Hence, while I agree with the fact that this would be difficult for application to all unknown particle types, I think there is quite a bit known and often assumed for dust relevant shape factors and densities. So the statement that "If actual particle densities were mostly > 1 g cm^{-2}, our [$n_{0.5}$] would be somewhat higher than if they would have been calculated as physical particle diameters" is unsatisfying. Without showing the size distribution, one does not know how particle numbers fall off with size. Could it amount to a factor of 2? For SD, I think it easily could. For SD especially, I think that shape and density factors are probably fairly well constrained, and a 542 nm aerodynamic diameter could easily be 380 nm. It is simply a misapplication of the parameterization to use aerodynamic diameter in it and compare to aerodynamic number concentrations. If you want to argue that aerodynamic diameter is more suitable for parameterizations, that is another matter. Using a relevant assumed shape factor (perhaps1.3) and density (perhaps 2.6) for SD in order to correct the parameterization to aerodynamic size and concentration space, this will move the D15 line to the right in the plots. How much? Using your data for SD number concentrations at two aerodynamic sizes and assuming that a linear slope exists between them (possibly not a good assumption, especially for estimated smaller size particle concentrations where the distribution may be steeper), I estimate a 2 factor concentration push of the D15 curve to the right (i.e., 2 per liter where you have plotted 1 per liter).

Thank you for the clarification and suggestion of a shape factor and a density. Using these values in combination with the size distributions measured during SD events pushes the D15 curve downward by a factor of around 2 on Fig. 3a with aerodynamical diameter as unit on the x-axis.

(P3L88-P4L92, **P3L87**) A physical diameter of 0.5 µm is equivalent to an aerodynamic diameter of 0.9 µm, assuming a particle density of 2.6 g cm^{-2} and a shape factor of 1.3 (Raabe, 1976), which are typical values for mineral dust particles. Similar transformations for observations not dominated by mineral dust would require information about densities and shapes of the main components of sampled particle populations, which were not available for our site and would require unsupported assumptions. Therefore, we chose

to show for all our observations the directly measured particle concentrations in terms of *aerodynamic diameter*. To use D15 parametrisation in our context, we corrected predicted [INP] for the difference between the aerodynamic diameter measured and the physical diameter used in Eq. 1 by multiplying $n_{0.5}$ in Eq. 1 by the ratio of particles with aerodynamic diameters > 0.9 µm (equivalent to 0.5 µm physical diameter) to particles with aerodynamic diameters > 0.5 µm, which we observed in Saharan dust dominated air masses during our campaign. The average value of this ratio was 0.59.

6) When you consider the above conservative estimate of how the D15 curve needs to be pushed to the right, and the fact that you should be using cf = 3, I would judge that your data for SD episodes are completely in line with D15 in Figure 3. I am less sure how to fix Figure S3, but perhaps that is fixed if the D15 number predictions are fixed. This means as well that the D15 parameterization grossly under-predicts INPs in other standard conditions. This makes total sense to me for situations where biological or other INPs dominate. Making these changes is simple, in my opinion, and then the D15 comparison only focuses on dust scenarios, and the other significant results in the paper (larger size relation to total INPs needed for this and possibly other sites, and the apparent role of biological INPs) remain unimpeded by this focus on a parameterization that does not account for them.

Using *cf* = 3, instead of *cf* = 1, pushes the D15 curve upward by a factor of three, while scaling D15 to the aerodynamic diameter pushes it downward by a factor of two (Fig. R1). Combined, the difference to the previous position of the curve is small (a factor of 1.5 upward).

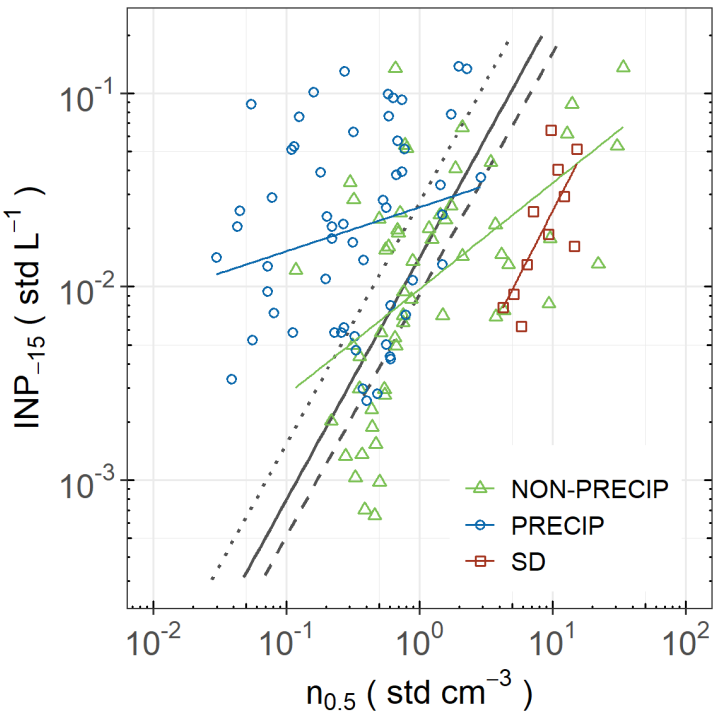
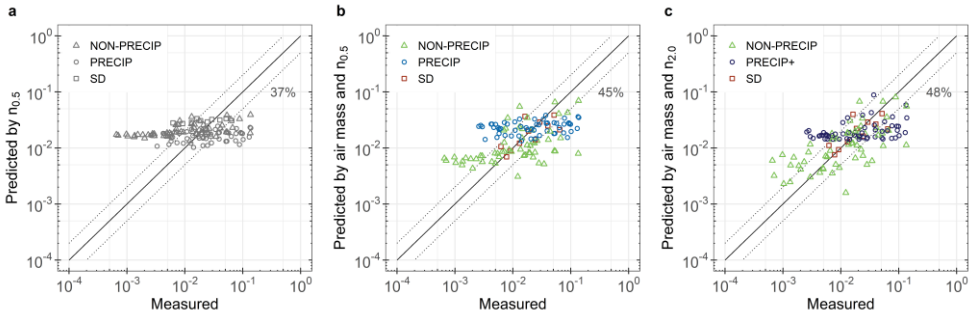


Figure R1: Same as Fig. 3a with the D15 parametrisation not scaled to aerodynamic diameter using two different calibration factors (cf = 1, dashed line; cf = 3, dotted line) and scaled to aerodynamic diameter using cf = 3 (continuous line).

Our comparison with D15 is now focused on SD air masses using different calibration factors. Fig. S3a of the first version of the revised manuscript was dropped because it unjustifiably applied D15 to air masses for which it was not developed. Fig. S3 is adapted accordingly.



(Figure S3, **Figure S3**). Measured and predicted cumulative concentrations of ice nucleating particles active at -15°C [INP_{-15}] (std L^{-1}) for (a) prediction based on a single trendline fitted through all data of aerosol particles with aerodynamic diameters > 0.5 µm [$n_{0.5}$], (b) predictions based on [$n_{0.5}$] and three different trendlines fitted through the data of PRECIP (blue circles), NON-PRECIP (green triangles), and SD (red squares) air masses, and (c) same as (b), but based on aerosol particles with aerodynamic diameters >2.0 µm [$n_{2.0}$]. Shapes in (a) are consistent with those in (b). However, they are coloured in gray as the prediction is independent of air mass classes. A range of a factor of two (dotted lines) about the 1:1 line (solid line) as well as the percentage of values lying within that range are shown in all panels.

Last, we added a few words to a sentence in the Conclusion, which was erroneously not that precise.

(P6L158; **P6L167**) The absolute value of additional INPs in precipitating air masses, versus non-precipitating air masses, seems to be independent of total aerosol concentrations.

References

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Schrod, J., Weber, D., Drücke, J., Keleshis, C., Pikridas, M., Ebert, M., Cvetkovi´c, B., Nickovic, S., Marinou, E., Baars, H., Ansmann, A., Vrekoussis, M., Mihalopoulos, N., Sciare, J., Curtius, J., and Bingemer, H. G.: Ice nucleating particles over the Eastern Mediterranean measured by unmanned aircraft systems, 17, 4817–4835, 2017.