Anonymous Referee #1

Marinou et al. show nicely the combination of field and laboratory work. Aerosol surface area concentrations derived from lidar observational data are used together with ice nucleation parameterizations derived from laboratory experiments to determine vertical profiles of aerosol-specific INP concentrations. The method is not know, but the authors included more state-of-the-art ice nucleation parameterizations and compared the INP number concentrations with offline analyzed filter samples taken with an UAV.

The comparison looks very promising. The authors show both immersion freezing and deposition nucleation nINP, although in the presented cases deposition nucleation would be very unlikely. It would be nice to see a follow-up study for a real deposition nucleation case.

[REPLY] We thank the reviewer for his/her careful reading, comments and suggestions, which we address in the following. With his/her suggestions, we believe that the new version of the manuscript is significantly improved. The author's replies along with the changes in the manuscript are listed below.

> Remark: The figure numbers and the page numbers in the referee comments are corresponding to the original manuscript. If not stated otherwise, figure and page numbers in the authors' answers are referring to the revised, marked-up manuscript version (showing the changes made) which can be found attached to this answer.

General comments:

The manuscript is well structured, however some paragraphs are unnecessarily long, e.g. description of the differences of the parameterizations. Your focus is the case 20-22 April. So in my opinion you should shorten section 4.1 (description of the other cases) or discuss the other cases similarly.

[REPLY] We moved the description of the other cases from section 4.1 to the beginning of section 4 (so now all the cases are together) and we shorten their description. The new text is provided in the reply of comment nr.11 below. Additionally, we include in the text the arguments that characterize the event of 20-21 April as our golden case and the reason that we separate that case from the rest cases by adding:

In page 12 line 24 "The pure dust event on 20 to 21 April 2016 is considered the golden case of our dataset, as it has been observed simultaneously with the PollyXT lidar, the UAVs and the A-Train satellites. Additionally, it is the only pure-dust event of our dataset where we have simultaneously good lidar observations and in-situ INP measurements."

In page 16 line19 "The sample of 21 April … This sample is used in order to evaluate the performance of the n_{INP} lidar estimates in a pure dust case, where (i) the errors originating from the first step of our methodology (separation in dust and non-dust aerosol components) are small ($\sim$ 30%) and (ii) the uncertainties induced from the D10 and U17-(soot) parameterizations are minimum."

The manuscript is well written, but I would propose to the authors going carefully through the paper and eliminate the typos and grammar error (some of them listed in the Technical comments section).

[REPLY] We read carefully the paper and eliminate the typos and grammar error that we find along with the ones that are listed in the technical comments section. Thank you very much for this.

Specific comments:

1. Abstract A major point in your work is the comparison with the FRIDGE INP measurements from filters taken with a UAV. However, this is not mentioned in the abstract.

[REPLY] We thank the reviewer for this comment. We add tis information in the abstract.

New version: page 1, line 7: "Here, we assess the feasibility of this new method for both ground-based and space-borne lidar measurements, using in-situ observations collected with Unmanned Aerial Vehicles (UAVs) and subsequently analyzed with the FRIDGE (FRankfurt Ice nucleation Deposition freezinG Experiment) INP counter from an experimental campaign at Cyprus in April 2016."

2. p. 2 l. 19 "about 1 in a million aerosol particles act as INP" This statement is well known, but I would prefer a reference.

[REPLY] We add the reference of Nenes et al. (2014): Nenes, A., Murray, B., and Bougiatioti, A.: Mineral Dust and Its Microphysical Interactions with Clouds, In Knippertz, P., and Stuut, J.B., Mineral Dust: A Key Player in the Earth System, pp. 287-325, Springer, ISBN 978-94-017-8977-6, 2014.

New version: page 2, line 24: ".. (about one particle in a million act as INP; Nenes et al. (2014)).."

3. p. 3 l. 1-3 This finding is not limited to field studies.

[REPLY] You are right. We corrected the phrasing of this sentence.

New version: page 3, line 8: "Observational studies have shown that immersion freezing dominates at temperatures higher than -30°C, while deposition nucleation dominates below -35°C (Ansmann et al., 2008, 2009; Westbrook et al., 2011; de Boer et al., 2011)."

4. p. 4 l. 5 As far as I see, this listing is general. If so, than you might add the review by Murray et al. (2012) for another soot (immersion freezing) parameterization.

[REPLY] We add the Murray et al. (2012) parameterization in the sentence.

5. Table 1 First, to increase consistency you should use either K or degC. Second, the parameterization function U17-imm dust is wrong, if T is in K (as in the other equations)!

[REPLY] We thank the reviewer for this comment. We corrected the U17-imm dust formula in the table. Now in all the equations T is in K.

6. p. 4 l. 33-35 This statement is true, but D15 uses for its parameterization next to lab data also field data and therefor, the explanation for the discrepancy is not appropriate.

[REPLY] I cannot find the explanation for discrepancy the reviewer refers to in the text in page 4 line 33-35 (or around). Please send me the specific extract from the manuscript. In

the meanwhile, in the new version we emphasized the use of filed data for the D15 parameterization and we rephrased the discussion on the S15 enhanced freezing efficiency, so we consider the sentences more clear and accurate.

New version: page 4, line 35: "Additionally, the parameterization of DeMott et al. (2015) (D15) (Table 1; Eq. 2) addresses the immersion and condensation freezing activity of natural mineral dust particles based on laboratory studies using the continuous flow diffusion chamber (CFDC) of the Colorado State University's and **field data from** atmospheric measurements in Saharan dust layers."

New version: page 5, line 11: "… S15 (Table 1; Eq. 4) was based on dust samples from Arizona, which were treated (washed, milled, treated with acid) and are much more ice active than **natural** desert dusts **particles** on average. Although S15 parameterization was based on **"treated"** dust samples which usually show an enhanced freezing efficiency, it is used in the NMME-DREAM model (Non-hydrostatic Mesoscale Model on E grid, Janjic et al. (2001); Dust REgional Atmospheric Model, Nickovic et al. (2001); Pérez et al. (2006)) for INP concentration estimations (Nickovic et al. , 2016). For this reason, it is included in this work."

7. p. 5-6 First, the ordering is confusing, because the two nucleation mechanisms are mixed. Second, for the reader community a less technical description of the parameterizations would be valuable. It is obvious that soot and dust have a different ice nucleation behavior. I would suggest discussing the differences of the parameterizations in terms of the future outcome in your study. That means, when S15 shows a significant higher activated fraction then you would expect that the number of INP is much higher than for the U17-dep dust. However, the error discussion is very good.

[REPLY] we rephrase the section according the instructions of the reviewer. The new section is:

New version: page 5, line 30: "Figure 1 provides an indication of the relative differences of the observed n_{INP} in nature for immersion (right) and deposition (left) modes and in relation with the different aerosol compositions by showing a summary of the different nINP parameterizations. Specifically, the plot shows the fraction of the ice-activated particles ($f_i = n_{INP}/n_{50,dry}$) for desert dust (dark blue, orange, red, light blue), continental (green) and soot (black). The particle concentrations used here, are derived assuming an extinction coefficient of 50 Mm^{-1} for each of the different aerosol types (dust, continental, soot). The shaded areas take into account a range of the extinction coefficient from 10 Mm^{-1} (lower limit) to 200 Mm^{-1} (upper limit). The error bars mark the cumulative error in f_i that results from the uncertainty in the lidar observations and their conversion to mass concentration as well as from the errors in the respective parameterizations. An overview of the typical values and the uncertainties used for the error estimation in this study is provided in Table 2. The deposition nucleation estimations in the left panel of Figure 1 are provided for ss_i = 1.15 (solid lines) and ss_i = (1.05, 1.1, 1.2, 1.3, 1.4) (dashed lines) to give a perspective on the range of possible values. Note here that although the immersion parameterizations were obtained using measurements at the temperature ranges of [-30, -14]°C (U17-imm, dust), [-35, -21]°C (D15, dust), [-34, -18]°C (U17-imm, soot) and [-35, -9]°C (D10, continental), they are extrapolated herein to extend over the immersion-freezing temperature range (dashed part of the lines in the immersion mode chart).

Figure 1 (left panel) shows that, for deposition mode, the dust ice-activated fractions from S15 are several orders of magnitude higher than those of U17-imm (e.g. 4 orders of magnitude at -40°C and ss_i = 1.15%). Furthermore, the deposition 5ice-activation fraction of dust and soot (from U17-dep) differ significantly with soot being more active than dust for T <-38°C (up to 2 orders of magnitude) and dust being more active than soot for T >-38°C (up to 4 orders of magnitude).

Figure 1 (right panel) shows that, for immersion mode, the dust ice-activated fractions obtained from D15 are one order of magnitude lower than those calculated with U17-imm. Laboratory ice nucleation measurements and corresponding instrument inter-comparisons, have shown that at a single temperature between two and four orders of magnitude differences are observed as a result of the natural variability of the INP active fraction (DeMott et al., 2010, 2017) or the use of different INP counters (Burkert-Kohn et al., 2017). Hereon, we consider D15 and U17-imm as the lower and upper bounds of the immersed n_{INP} estimations for dust INP populations. Figure 1 (immersion mode panel) illustrates the dust activation increase of up to six orders of magnitude within the mixed-phase temperature regime (−15 °C to −35 °C). For a 5 °C decrease, $n_{d,INP}$ increases by about one order of magnitude. Moreover, we see that at T < -18°C the immersion freezing desert dust ice activation (D15) is higher than the continental one (D10) while this changes at T > -18°C. On the contrary, soot (U17-imm) has always lower f_i than dust (from either D15 or U17-imm). The ice-activated fractions of continental (D10) and soot (U17-imm) aerosols have a relative difference that is always less than 60% at T < -18°C. At higher temperatures they diverge with continental fi to exceed the soot one by one order of magnitude at T > -11◦C."

8. p. 7 l. 3 "(…) several in-situ instruments were operated" for what? What did they measure? Be more specific or remove that, because you do not use these instruments.

[REPLY] Thank you very much for this suggestion. We remove that part from the sentence, as indeed we do not use any of these data here.

New version: page 7, line 25: "An Aerosol Robotic Network (AERONET, Holben et al. 1998) sun photometer was located at the Cyprus Atmospheric Observatory of Agia Marina Xyliatou (35°02'19"N, 33°03'28"E, 532 m asl, 7 km west of the UAV airfield)."

9. p. 8 l. 10 Can you give a reference for the SAMUM experiment?

[REPLY] For a general reference of the SAMUM1 and 2 experiments, we add the reference: Ansmann, A., Petzold, A., Kandler, K., Tegen, I., Wendisch, M., Müller, D., Weinzierl, B., Müller, T., and Heintzenberg, J.: Saharan mineral dust experiments SAMUM-1 and SAMUM-2: what have we learned?, Tellus B, 63, 403–429, doi:10.1111/j.1600- 0889.2011.00555.x, 2011b.

New version: page 9, line 5: "… This assumption has been validated against airborne in-situ observations of the particle size distribution during the Saharan Mineral Dust Experiment (SAMUM; Ansmann et al. (2011b)) in Morocco."

10. p. 8 l. 11 This number was not given in percent, right?

[REPLY] Yes, you are right. We corrected it in the manuscript.

New version: page 9, line 7: “The correlation drops to ≈0.85±0.10 for urban environments based on ground-based in-situ measurements of particle size distributions at the urban site of Leipzig (Mamouri and Ansmann, 2016).”

11. Section 4 and 4.1 In the very first part of Section 4 you describe detailed the case of 21 April, but not the other cases. These cases are discussed in Section 4.1. This is confusing for the reader.

[REPLY] We thank the reviewer for his comment. He is right, so we move the discussion of all the cases in the beginning of Section 4 (from page 11 line 32 to page 13 line 12), and in the new version only the comparison between the UAV-measured and lidar-derived concentrations are discussed.

12. p. 11 l. 27-28 What was the height for 5 April?

[REPLY] We add the following sentence in the manuscript:

New version: page 15, line 2: “These measurements correspond to heights above 0.5 km on 5th of April.”

13. p. 11 l. 34, Figure 7 Ok, but there is a deviation from the 1:1 line especially for the high concentrations (or case 9 April). Do you have an explanation or can you comment on that?

[REPLY] We add the following explanation in the manuscript:

New version: page 15, line 6: “On 9 April we observed the highest differences between the lidar-derived and in-situ-measured $n_{250,dry}$, which may be attributed to the ∼1 hr time difference between the in-situ sampling and the lidar retrieval (limitation due to mid-level clouds as discusses already). Nevertheless, the case is included here, as it represent the strongest dust event observed during the campaign.”

14. p. 13, Figure 8 and 9 The discussion of the two figures is quite similar and at some point you repeat the findings. Maybe you can shorten this part.

[REPLY] In the new version, we have merged the discussion of these 2 figures (Figure 9 and 10 in the new version). The discussion is shorter in some extend and there are no repetitions that were present before.

New version:

page 17, line 9 – line 18: “Figure 9 (b) and Figure 10 (b) shows …”

page 17, line 19 – page 18, line 12: “In Figure 9 (b) and Figure 10 (b) we see that …”

15. p. 13 l. 18ff You did not this detailed discussion for deposition nucleation. Be more consistent.

[REPLY] We tried to be more consistent when discussing the immersion and deposition modes. In the new version, the deposition nucleation results are discussed in 10 lines (page 14, lines 19 – 28 in the final not marked up version) and the immersion/condensation results in 32 lines (page 14, line 29 – page 15, line 25 in the final not marked up version). The reason for the remaining difference is attributed to three things:

1. From the 2 existing deposition parameterization (S15, U17), we initially know that the S15 one is not good enough for natural desert dust (as it is based on treated dust samples with

modified ice activity) but we include it anyway for completeness purposes as it is currently used in the BSC-Dream model. On the other hand, the parameterization of U17-dep provided excellent agreement with the in-situ measurements; hence, we do need any discussion on disagreement with the in-situ (as there is none).

2. For immersion mode, there are many parameterization in the literature available (D15, D10, U17) which are based on natural aerosol measurements, but they provide different INPC results. Additionally, the differences observed, even for the same parameterization, varied a lot (from identical up to 3 orders of magnitude different than the in-situ INPC - when the samples are analyzed in different temperature). In the manuscript, we discuss these differences and the possible sources of discrepancies and errors of the in-situ measurements (FRIDGE is widely used for immersion measurements but it was originally constructed for deposition nucleation measurements and hence the deposition IN measurements are more accurate).

3. In the discussion of immersion/condensation INP estimates, we provide indicatively some INPC values of the in-situ measurements and the lidar retrievals for the case of 21 April, as is later on discussed in detail and in comparison with cloud n_{ICE} observations in section 4.3. The relevant temperatures in this case are <-35°C hence only the immersion INP estimates are interesting to be mentioned.

16. p. 15, Figure 10 From the campaign, are there temperature and/or relative humidity measurements available e.g. from radiosondes? From the WRF temperature profiles, you could argue that deposition nucleation will not be the case for your study. Furthermore, you could add the approximate cloud base and top height in Figure 10.

[REPLY] The WRF modeled profiles used are assimilated with the NCEP global reanalysis dataset. We have included this information in section 3.3 (page 9, lines 18 – 25).

Also, we added the following argumentation in the discussion of this case (page 19, line 20): "From the WRF and MERRA-2 assimilations we see that T < -35°C in heights up to 7.8 km agl, which indicate that the immersion freezing mechanism is dominant in this case and that the deposition nucleation mechanism is not significant."

Furthermore, we add a new figure where we are indicating the cloud boundaries in this event next to the n_{INP} values: new Figure 13.

17. Summary section The conclusion are very short. Maybe you can discuss in more detail what improvements you or the community can do to improve the outcome, e.g. collocated temperature/ humidity profiling for calculating the INP concentration at real conditions, or combined in-situ ice concentration measurements.

[REPLY] We included the following discussion in the conclusions:

New version: page 21, line 32:" A further step for improving the lidar-derived INP retrievals and investigating the different parametrizations used is by conducting dedicated studies with collocated lidar measurements and additional temperature and humidity profiling in order to calculate the INP concentrations at real conditions, and the combination of the retrieved n_INP with airborne in-situ ice concentration measurements."

<u>Technical corrections:</u>

1. **p. 1, l. 6 Either "(…) lidar measurements with a INP efficiency (…)" or "(…) lidar measurements with INP efficiency parameterizations (…)"**

[REPLY] Corrected as "(…) lidar measurements with INP efficiency parameterizations (…)"

2. p. 1 l. 12 14 agrees

[REPLY] Corrected

3. p. 1 l. 12 nINP not yet introduced

[REPLY] Changed to "INP concentrations (n_{INP})"

4. p. 2 l. 6 "Our analysis" either has shown or shows "that (…)"

[REPLY] Corrected to "shows"

5. p. 2 l. 8 gives

[REPLY] Corrected

6. p. 2 l. 8 Neither n250,dry nor Sdry introduce

[REPLY] The n250 was introduced in the previous paragraph and the Sdy in this line. But indeed, they were not comfortably understood while reading this part. We change this sentence and include their definitions in parentheses next to the symbols.

New version: page 3, line 22: "Lidar measurements can provide profiles of $n_{250,dry}$ (the number of aerosol particles with dry radius greater than 250nm) and S_{dry} (the aerosol particles dry surface area concentration) related to mineral dust, continental pollution and marine aerosol..".

7. p. 2 l. 30 citation style in the brackets

[REPLY] Corrected

8. p. 3 l. 32 UAV comes first here, write out in full

[REPLY] done

9. p. 4 l. 3 citation style

[REPLY] Corrected

10. p. 4 l. 16 AIDA comes first here, write out in full

[REPLY] Corrected to "Aerosol Interaction and Dynamics in the Atmosphere (AIDA) cloud chamber"

11. p. 4 l. 27 "need to be transferred"

[REPLY] Corrected

12. p. 4 l. 32 "(…) from Arizona, which have been (…) and are much more (…)"

[REPLY] Corrected

13. p. 5 l. 5 devices

[REPLY] Corrected

14. p. 5 l. 7 citation style

[REPLY] Corrected

15. p. 5 l. 13 shown

[REPLY] Corrected

16. p. 6 l. 4 desert

[REPLY] Corrected

17. p. 6 l. 10, 13, 16 5 degC

[REPLY] Corrected

18. p. 9 l. 28 "(…) and the Arabian Peninsula to the Eastern Mediterranean (…)"

[REPLY] Corrected

19. p. 11 l. 28 seems

[REPLY] Corrected

20. p. 12 l. 24 "(…) microscopy, which shows that (…)"

[REPLY] Corrected

21. p. 13 l. 35 than instead of that

[REPLY] Corrected

22. Figure 2 right figure Sdry has a wrong unit

[REPLY] Thank you very much. We corrected the unit.