Referee report on "Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater" by Xianda Gong et al.

The manuscript presents an analysis of filter and water samples for their content of ice nucleating particles (INPs). Samples were collected at the Cabo Verde Island as part of the MarParCloud project. What makes this study unique is that samples were simultaneously taken at sea and cloud level, and include filter samples of ambient air as well as cloud water and sea water samples. By comparing the INP content in these different compartments of the environment the authors infer the contribution of particles of certain origin and characteristics to the atmospheric INP population.

There are several points that need to be improved -- these are listed in detail below. Once these are addressed, the paper should be published in ACP.

Specific comments

1) P.1 ln.14f Attributing the difference in N_{INP} from PM10 and PM1 samples to biological particles only based on the temperature of activation is speculative and over-reaching, eg. super-micron mineral dust particles can also cause ice formation at -10°C (see Hoose and Möhler, 2012). Are there indications against mineral dust particles being responsible for the higher activity? Were elevated N_{INP} observed during dust events?
2) P.1 ln.18 Same as above. Most particles >1um are probably activated as CCN. There is no evidence for a biological origin of these INPs.
3) P.2 ln.7 In addition to homogeneous ice nucleation, heterogeneous ice nucleation by deposition nucleation is active also below -38°C. As the sentence is written it indicates that only homogeneous nucleation would be active below -38°C, which is incorrect.
4) P.2 ln.9 Are all droplets aqueous solutions?
5) P.2 ln.12 If there is a special aspect to the -20°C reported in Augustin-Bauditz et al., 2014 it should be mentioned. If not, the "below -15°C" from Hoose and Möhler, 2012 already includes -20°C and the Augustin-Bauditz et al., 2014 reference can be omitted. According to Fig. 3d in Hoose and Möhler, 2012, super-micron mineral dust particles can be active INP already below -10°C and not -15°C.
6) P.2 ln.17 The results in Boose et al., 2016 suggest desert dust to nucleate ice mostly below -25°C, especially airborne samples and the study shows differences among samples from different regions. This seems to contradict what is stated in ln.12-16. This section of the introduction should be revised, distinguishing mineral dust from desert dust studies and motivating the relevance of desert dust for the results presented here.
7) P.2 ln.26 Specify what is meant by "differences in desert sources".
8) P.2 ln.28 Is N_{INP} at ground level lower because the dust loading was lower at ground level?
9) P.2 ln.32f This is incorrect. Bigg, 1973 suggested that INPs "are transported from a distant land source, or from a stratospheric source, and brought to sea level by convective mixing." Schnell and Vali, 1976 suggested a marine source could explain the observations of Bigg 1973.
10) P.3 ln.2 What further evidence? This sentence seems to refer in a more general tone to the same study as the sentence before. Please clarify.
11) P.3 ln.10f DeMott et al. 2010, 2015 suggest a correlation of N_{INP} and the concentration of particles above a certain size. However they do not specify that only a fraction of large particles would act as INPs. Double check.

12) P.3 ln.20 The deduction that most biological INPs occur together with their original carrier appears incomplete. Clarify.
13) P.4 first paragraph of Sec. 2.1.1. If the CVAO station is located on the northwest shore (ln. 11) of the island and wind direction is from the northeast (ln. 17), does this mean air first crossed the island before reaching the station? I think the CVAO is located on the northeast shore.
14) P.5 ln.16 It is mentioned in Sec. 2.1.1. that measurements took place during 31 days. Why where only 17 or 19 filters collected? How long was the sampling period per filter?
15) P.6 ln.29 By "brightness change" do you mean a change in transmitted light due to a change in opacity when droplets freeze?
16) P.7 ln.8f Can you explain how the assumption of a Poisson distribution influences the result of Eq.1?
17) P.7 sec.2.3.1. The principle of determining the cumulative INP concentration as well as the difference between INDA and LINA experiments are not very well explained. The explanation should mention that all wells contain multiple INPs but only the most active one (active at the highest temperature) causes freezing. The probability to find an active INP at a certain temperature increases with sample volume.
18) P.7 Sec.2.3.2 Provide the equations you used for this calculation and some values to inform the reader how large statistical error and background are.
19) P.7 ln.18 It is not explained previously that washing water was used or how it was prepared.
20) P.7 ln.18 How was the information about the number of INPs per well obtained?
21) P.7 ln.20 A short explanation of the method from Agresti and Coull, 1998 would be helpful here.
22) P.8 ln.3f Can you provide a range of the derived freezing point depression for the samples. Was the freezing point depression experimentally confirmed, eg. by measuring the melting point depression?
23) P.8 ln.9 The description of n_s is imprecise. N_s gives the number of ice active sites per surface area, here the surface of all aerosol, ice active or not. Revise.
24) P.8 ln.20-26 This section is speculative and it is not clear why this comparison is relevant for the present study. Clarify.
25) P.8 ln.27 Add an introducing sentence mentioning that the following analysis is done to compare N_{INP} found in SML and ULW at Cabo Verde and explain why enrichment/depletion could be expected. Could INP in the SML originate from settling aerosol?
26) P.9 Fig.1 Why were samples 6, 7, 8, 10 and 11 not used? Instead of comparing to Wilson's data from a different environment, a direct comparison of SML to ULW by plotting both data on top of each other might show more clearly that N_{INP} are the same between the two.
27) P.9 ln.2 Clarify that you refer to the temperature at which N_{INP} was determined in the drop freezing experiment. As is, it could be misunderstood as water temperature during sampling.
28) P.9 ln.3 Can you provide an explanation for the variation in EF with temperature? Does the interpretation change when considering the confidence interval of N_{INP}? Do an error propagation of Eq. (5) and estimate the error in EF (should be included in Fig.2).
29) P.9 ln.8 Shouldn't SML thickness be related to concentration of dissolved organic matter? Explain why SML at Cabo Verde is larger than the SML in the Wilson et al., 2015 study even though conditions are oligotrophic.
30) P.10 Fig.2 add error estimation of EF.
31) P.10 ln.9-11 What could be the reason for the variation between samples and why is the range of variation consistent to measurements at other locations? Could the number of samples or sampling duration determine the range of variation?

32) P.10 ln.12f Testing the heat sensitivity of the 3 samples could substantiate the interpretation that biological particles are responsible for the enhanced N_{INP}.
33) P.10 ln.14ff. This paragraph is difficult to follow. Do you mean above -16.8°C concentrations within 2°C of each other are correlated? Looking at the data in Fig.3, N_{INP} seem to change very little in 0.1°C steps, but are different between individual measurements. What is the actual regression model for which you report R^2 and p value? What data was used for the regression? Check the statistical power of correlating only few data points. Looking at the data in a differential spectrum (see Vali, 1971) might be a better method to identify temperatures where INPs of different origin become active.
34) P.11 ln.5 The dataset is by far not large enough to construct a robust pdf. The result in Fig.4 is vastly dependent on the choice of intervals to bin the data. The given pdf is therefore not suitable to perform any data analysis as the result could depend on the binning of data.
35) P.12 Fig.4 I suggest to remove Fig.4. It is not illustrating new information that is not already contained in Fig.3 and Fig.5. If you chose to keep Fig.4 check y-axis, the area under the pdf should be 1 or 100%.
36) P.12 ln.6ff Explain how the values in this paragraph were derived. I assume you compare the $N_{INP,PM10}$ to $N_{INP,PM1}$ from filters collected during the same time period and take the ratio?
37) P.13 ln.3ff Last sentence of this paragraph is speculative and repeating for the 6th time in this manuscript that high temperature activity of PM10 filter samples could be due to biological particles. As this seems to be a central point in your interpretation of the data I strongly recommend to experimentally test the heat sensitivity of N_{INP} (eg. following the procedure described in Joly et al., 2014) to support that biological particles are causing the mentioned difference.
38) P.14 ln.1 The difference in N_{INP} (shown in Fig.6 (b)) is clearly visible above -20°C, not only above -17°C.
39) P.14 ln.3f Is there evidence for a substantial fraction of droplets below 10um? Even though no direct observations are available from MV, observations in similar environments could help this discussion. Measurements of orographic cloud droplet distributions e.g. from Hawaii showed a bimodal droplet size spectra with both modes >10um (Squires, 1958).
40) P.14 ln.5 This speculation is repeated several times throughout the paper but no evidence to support the biological nature of these INPs is presented. Either conduct heat sensitivity experiments and/or provide electron microscope images of large biological particles on the filters to demonstrate that this is a plausible interpretation, or delete the statement.
41) P.15 ln.4ff In some cases over 100% difference in Na^+ and Cl^- concentration between the present study and Gioda et al., 2009 seem large and not comparable. In contrast to what other values do the authors think concentrations are comparable?
42) P.16 ln.13ff Couldn't F_{cloud_air} be estimated directly from the water collection rate of the CASCC2? This would reduce the uncertainty for the estimation of N_{INP}.
43) P.17 ln.1f Does this range include the error estimation from the INP experiment? The two uncertainties (in F_{cloud_air} and N_{INP} in cloud water) should be combined by error propagation when deriving the range of $N_{INP,air}$.
44) P.17 ln.3 The uncertainty range spreads over 2 orders of magnitude while the N_{INP} cover 4 orders of magnitude. "general agreement" seems to have limited meaning here. The sensitivity on d_{drop} when calculating $N_{INP,air}$ from water samples determines the result. As already suggested above, could the amount of collected cloud water be used to determine F_{cloud_air} or to constrain the d_{drop} range? Alternatively LWC can be estimated from the CASCC2 collection rate (Sec. 2.3. in Demoz, 1996) for different drop size distributions (that could

come from the literature eg. Sqires, 1958). Another option to estimate LWC might be to use the NaCl content in cloud water and air as a tracer, similar to the method applied in Sec.3.4.

45) P.17 ln.5 Instead of the collection efficiency at 3.5 um, it would be more useful to know the collection efficiency above the cut-off of the PM10 inlet of the filter sampler, where the droplet fraction not collected by the filter sampler but the cloud water collector should be found. According to Demoz, 1996 collection efficiency above 10um should be >80%. The high collection efficiency above 10um does not support the given explanation for a difference in N_{INP} from filter and cloud water samples which is provided at the beginning of the paragraph (ln.3ff). Revise.
46) P.18 Eq. (7) and Fig.9 An error estimation for N_{INP} from sea spray by error propagation of input variables in Eq. 7. Include error estimate in Fig.9.
47) P.18 ln.7 Did you use the individually measured NaCl concentration for each sampling period or the median to calculate the INP concentration in air? What is the range of the NaCl ratio on the right hand side of Eq.7?
48) P.18 ln.10f Related to the previous comment. N_{INP} at Cabo Verde and in the Arctic should be the same only if the NaCl ratio in Eq.7 is also the same. Alternatively, this highlights that the result of Eq.7 is largely insensitive to the NaCl ratio. This should be clarified.
49) P.19 ln.1-12 It is unclear why enrichment of OC in SML is discussed here as no connection to N_{INP} has been established. I recommend to delete this paragraph. All that can be said is that airborne N_{INP} are higher than whatever N_{INP} could have originated from the ocean.
50) P.20 Fig.10 I suggest to include n_s for all temperatures covered by your experiments and for filter, water, CVOA, MV separately.
51) P.20 ln.9 Fig.10 should be motivated by stating what the expected n_s are (SSA or dust) and then argue that the available n_s parametrizations are not representative for Cabo Verde. It might be not surprising that n_s parametrizations that are based on measurements in other environments do not capture the situation at Cabo Verde. Additionally, specify which data of "our data" is shown in Fig.10. Is it filter, water, CVAO or MV? As suggested in the previous comment all of these datasets could be of interest.
52) P.21 ln.5f Here, the authors could suggest future directions, eg. regional, seasonal n_s parametrizations or parameterizing N_{INP} directly from field observations without employing surface area specific activity.
53) P.21 ln.19f Repetition of sentence from p.10 ln.12f. What could be the origin of these super-micron biological particles? Doesn't the evidence in this paper rather point to super-micron mineral dust?
54) P.21 ln.24 Provide evidence for biological particles or include dust as a possible source.
55) P.21 ln 26-27 Either, add figures showing both N_{CCN} and PNSD at the two locations during cloud events and non-cloud events and provide difference in PNSD where the CCN active INPs are found, or, point the reader to Fig.8 in the companion paper. Was there a trend in N_{INP} related to the particle types indicated in Fig.8 of the companion paper?

Technical corrections

1) p.1 ln.4f SML and ULW might be sources of INPs, but sea and cloud level are compartments of the atmosphere where N_{INP} are measured, not sources. Rephrase.
2) P.1 ln.7 When mentioning "temperature" be specific in which system the temperature was measured. Here: "trends of EF with temperature." Temperature of what? Sea water, ambient or in the INP experiment?

3) P.1 ln.8 Same as above. “at any particular temperature” could be understood as if sampling was conducted at different temperatures. The authors should be more specific and say: the temperature to which samples were exposed to in ice nucleation experiment.
4) P.2 ln.10 Freezing is not the same as ice nucleation. Immersion freezing refers to an ice nucleation mechanism rather than the freezing process. Rephrase.
5) P.2 ln.15 Replace “more effective” with “more active” instead.
6) P.2 ln.22 Do you mean North African desert?
7) P.2 ln.27 Replace “ice nucleating properties” by “N_{INP}“
8) P.3 ln.1 Replace “INPs” with “the ice nucleation activity”.
9) P.3. ln.22 Add: assuming that most INPs activate as CCN.
10) P.3 ln.23 Specify: “in rain samples”
11) P.3 ln.31 Replace “for INPs analysis” with “to measure N_{INP}“
12) P.5 ln.4-6. Repetition of “specially designed”. Delete in line 4-5.
13) P.5 ln.12 Is there something special about the Digitel filter sampler from the reseller Walter Riemer Messtechnik? If not the manufacturer should be referenced instead.
14) P.5 ln.13 Move (Munktell, MK 360) to after “filters”.
15) P.8 ln.12 Please add units to variables.
16) P.8 ln15 Mention that this sentence refers to individual samples and starting from ln.17 variation between the 9 samples is discussed.
17) P.8 ln.26 Instead of “This” start the sentence with “The low biological activity in the SML around Cabo Verde”
18) P.9 Fig.1 Use the same y-axis scale for SML and ULW and include gridlines to facilitate comparing SML to ULW. Consider plotting the data on top of each other.
19) P.9 ln.7 Specify if you refer to sampling, or INP experiment technique.
20) P.9 ln.10 Delete “the”
21) P.10 ln.7 “contribute” instead of “contributes”
22) P.10 ln.7 Replace “few” with “two”
23) P.13 Fig.5 Check unit of y-axis. Should not be %. Add to the figure caption what range is represented by the box and whisker of the boxplot. Due to the limited number of samples it would be better to just provide the range instead of a boxplot (which requires the assumption of an underlying distribution).
24) P.13 ln.12, 14, 17 Avoid vague qualifiers “more or less”, “only little”, “quite similar”, “mostly” and quantify instead.
25) P.13 ln.19 Add: “…obvious from Fig.6 that…”
26) P.14 Fig.6 Add (a) and (b) to the subfigures and add gridlines for easier readability. In the caption put the (a),(b) before describing the subfigure: “… MV PM_{10} filters during (a) less (cloud time fraction <10%) cloud effected periods and (b) highly…”
27) P.16 ln.25-28 Give the equation for this calculation.
28) P.17 Fig.8 First sentence in figure caption is incomplete. Replace “shown by” with “shown as”.
29) P.18 Fig.9 Caption: “error bars showing” instead “error bars show”
30) P.19 ln.26-28 Revise structure of sentence.
31) P.19 ln.31 Add: “… ice activity of super-micron mineral dust…”
32) P.20 ln.5 Add: “… associated with biological particles, but has also been observed for super-micron dust samples (Hoose and Möhler, 2012).”
33) P.21 ln.4 You could add: “… do not originate from sea spray, but are dominated by super-micron dust.”

34) P.21 ln.9 Instead of “thorough analysis” specify what kind of analysis is shown in the companion paper.
35) P.21 ln.13f Freezing experiments with the devices used for this study should give reliable data up to 0°C and not “roughly” from below -5°C.
36) P.21 ln.15f Revise after correcting Sec. 3.1.
37) P.21 ln.21ff Revise after correcting Sec. 3.2.1.
38) P.21 ln.25 “quite similar” should be put into perspective based on the limited number of investigated samples.
39) P.21 ln.30f Revise after correcting Sec. 3.3.2

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