Dear Paul,

Thanks for doing this review, for your thorough reading of our manuscript and for your positive comments. The points you raised were mindful and certainly helped improving this manuscript. Below, please find your original comments in blue and our responses in black. When referencing page and line numbers, we are always referring to the new versions of manuscript and SI, which are attached at the end. Concerning the literature cited in this answer to your review, we ask you to refer to the attached new version of the manuscript with tracked changes.

## General Comments

This is a very well executed, and fairly comprehensive study of ice nucleating particles in the Cape Verde region, especially novel in including measurements in all water and air compartments, and attempting to relate these meaningfully. Overall I had only few comments of significance, and the rest are mostly editorial notes. Specific questions/comments for addressing before publication are listed below.

## Specific Comments

1) Abstract, last sentence: I struggled to understand this sentence, although I think it is saying that unless there is an unusual SSA INP emission mechanism in the study area, the INPs cannot be from SSA. But the use of the phrase "unless there would be" seems to beg a question that I thought the papered endeavored to answer. As I read, it seems inconceivable. Or are you referring to situations that you did not measure? I suggest thinking about rewording this sentence.

We changed last sentence in the abstract such that this should be clear now:

“This latter conclusion still holds when accounting for an enrichment of organic carbon in super-micron particles during sea spray generation as reported in literature.”

2) Intro, page 2, line 8: A minor note, since it is not relevant for the main topic of this paper. It is difficult to encapsulate this discussion that seems to be required for every INP paper, but this statement does not reflect any role for INP at temperatures lower than -38 C, which is not the case.

Thanks for your comment. We changed it as:

"Ice crystals in the atmosphere can be formed either via homogeneous nucleation below −38 °C or via heterogeneous nucleation aided by aerosol particles known as ice nucleating particles (INPs) at any temperature below 0 °C."

3) Intro, page 2, line 28: Higher than ambient INP concentrations at ground level?

Yes, it is. We extended this sentence as:

"Schrod et al. (2017) found that mineral dust or a constituent related to dust was a major contributor to $N_{INP}$ of the aerosol on Cyprus, and $N_{INP}$ in elevated dust plumes was on average a factor of 10 higher than $N_{INP}$ at ground level, where the dust loading is lower."

4) Intro, page 2, line 29: I do not understand the meaning of, nor see the need for, the ending phrase of this sentence (i.e., : : :from the biosphere). It is clear that most INPs come from the biosphere, and the ocean source comes in the form of sea spray aerosol emissions. I favor being explicit.

We deleted this ending phrase "which would…".

5) Intro, page 2, line 33: Bigg suggested that INPs were contributed to at least some extent from marine emissions, in the data collected in that region at that time. His abstract statement reads that it is not feasible that they are "only of continental origin."

You are correct in that Bigg (1973) suggested that the INPs are not only of continental origin, but he does not mention marine sources at all. Instead, he argues that there should be a stratospheric source. As the second review suggested, the text in our manuscript was changed to:

"Based on a long-term measurement of INPs in the marine boundary layer in the south of and around Australia, Bigg (1973) suggested that INPs in ambient air were 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)."

6) Intro, page 3, lines 10-11: This is an oft-misinterpreted point. These papers parameterize INPs following this segment of the aerosol population, especially in the free troposphere, but the intention is to reflect INPs at all sizes. It is simply a hook to these concentrations, not intended to represent an actual "fraction" of them.

Thanks for clarifying this. We reworded this sentence.

"Simultaneous measurements of $N_{INP}$ and particle number size distributions were used to develop parameterizations in which $N_{INP}$ depends on a temperature dependent fraction of all particles with sizes above 500 nm (DeMott et al., 2010, 2015)."

7) Intro, page 3, lines 16: Perhaps add qualifier that these observations were in the Arctic "boundary layer". I bring this up a few times because many things could differ in the free troposphere and at the level of some colder clouds.

It was changed to:

"Creamean et al. (2018) also found that super-micron or coarse mode particles are the most proficient INPs at warmer temperatures in the Arctic boundary layer and they might be biological INPs."

8) Experiment and Methods, page 4, line 13: The filter sampler was truly sitting at ground level at MV? Or what was the elevation of the sample head? E.g., 1 m above ground?

The filter sampling was done using a Digitel filter sampler. The filter head was about 2 m above ground level. As mentioned in the paper, the height of MV is 744 m and the inlet height is 746 m. We added:

"… on the ground with the inlet 2 m above the bottom, …"

9) Experiment and Methods, page 5, line 19: Notes on Table S2. The volumes listed do not seem to work out with other information provided, and the header units seem wrong. First, is duration actually in minutes instead of hours? Also, is std volume (must be L, not L-1) for the 2 cm2 surface stated as taken? It does not quite make sense, since if this represented 1/100 of the volume flow of

500 lpm, then about 10 times this volume should have been represented. Please fix header units at least. And state in table description if volume is for the "punch" or the total filter. I will revisit this in the next comment.

Thanks a lot for discovering these errors.

The “Duration” should be in minutes. The “Total Volume” should be in std $m^3$ and “Volume Per Well” should be in std L. The volume is always given in standard temperature and pressure (0 °C and 1013.25 hPa).

10) Experiment and Methods, page 6, lines 24-25: So if I have it correct, this amounts to about 0.75 cm2 of the filter surface area (96, 1 mm punches). Yet this area is stated as 2 cm2 in the previous section. And these new numbers still do not seem to work out to give the sample volumes stated in the tables. I ask these things only if someone wanted to reproduce such work.

For the here presented measurements, only a small piece of 2 cm in diameter was used, from the much larger filter. From this small piece, then pieces with 1 mm in diameter were punched out. The area of these 1 mm needs to set into relation with the size of the overall sampled area. For the filters with diameters of 150 mm, the effective sampling area had a diameter of 140 mm. We made this clearer in the text:

“on 150 mm in diameter quartz fiber filters (Munktell, MK 360) with an effective sampling area of 140 mm in diameter.”

“…a circular piece of these filters of 2 cm in diameter was used from which then smaller pieces were punched out for the analysis (see section 2.2).”

11) Experiment and Methods, Section 2.3.2, page 7: Was background testing also done for the other collections, for example by rinsing clean plates use for microlayer sampling and by rinsing the bottles used for seawater collection? Just looking for a few words.

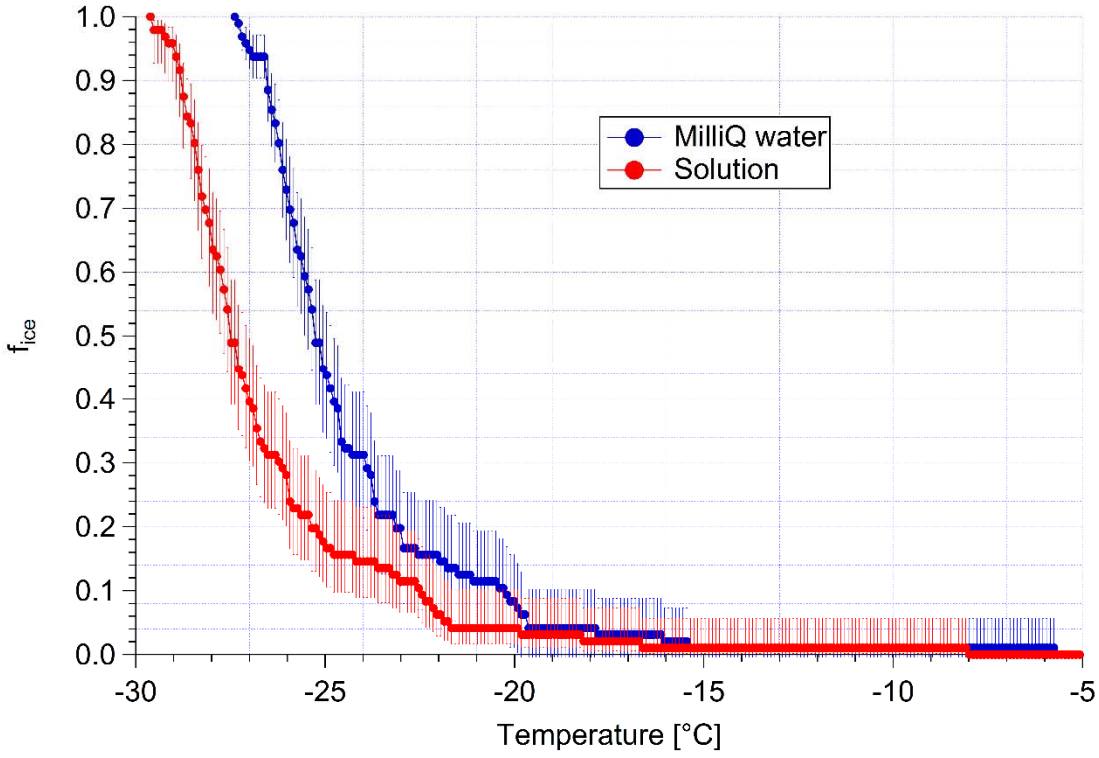
We tested the MilliQ water at Cape Verde and it was as clean as MilliQ water at TROPOS. But we did not test the MilliQ water after washing these glass plates and the containers in which the samples were stored. But we at least explicitly say now, at the end of this section:

“For those samples that were already collected in a liquid state (ULW, SML and cloud water), a background correction was not done.”

12) Experiment and Methods, Section 2.3.3, page 8: I am curious if this calculation of freezing point depression was checked for validation, by for example diluting a seawater sample?

We did not test the freezing point depression previously, but did it now. Since the seawater samples are no longer available, we tested the freezing depression of a pure sodium chloride solution.

We dissolved 0.72 g sodium chloride in 20 mL MilliQ water to get a solution with a salinity similar to that of the SML and ULW samples. The frozen fraction ($f_{ice}$) of MilliQ water and of this sodium chloride solution are shown in the figure below. The error bars show 95% confidence intervals of $f_{ice}$. Due to large measurement uncertainties for the first frozen droplets, the freezing point depression should rather be determined from temperatures below approx. −25 °C, where, indeed, a freezing point depression temperature about ~2.2 °C was observed. It is therefore acceptable to use a freezing point depression of 2.2 °C in this study.



13) Experiment and Methods, Section 2.4, page 8: When introducing surface active site density (the terminology I am used to it being referred to as), it could be good to mention already the fact that when applying it to the total aerosol distribution, this artificially assumes that all particles are the same INP type in the contained surface area. This is distinct from a laboratory scenario of generating a specific aerosol, and so will fold in all of the influences present in ambient air.

We followed your suggestion and extended this part as:

"For cases where a single type of aerosol, such as one type of mineral dust, is examined in laboratory studies, $A_{total}$ can be the total particle surface area. However, when field experiments are done, using the total particle surface area of the atmospheric aerosol assumes that all particles contribute to INP and have the same $n_s$, while the vast majority of these particles will not even be an INP. On the other hand, singling out the contribution of separate INP types in the atmospheric aerosol and relying $n_s$ only to them by using their contribution to the total surface area is at least demanding if not often impossible. This has to be kept in mind when interpreting heterogeneous ice nucleation in terms of $n_s$"

14) Results, page 13, lines 5-6: If bio-INPs, the size range is a bit unexpected for marine bacteria, which tend toward micron or less sizes I think. This might support that the bioaerosols are coming from long distance. It also strikes me that a mention for future work might be to include a collection at >2.5 microns, as one wonders about details of the INP size distribution.

We followed your suggestion and added:

"This suggests that these biological INPs might originate from long-range transport, as marine biological INPs were usually reported to be submicron in size (Wilson et al., 2015, Irish et al.,2017). The contribution of SSA to INPs will be discussed further in section 3.4."

15) Results, page 13, line 12 and Fig. 6: It is not optimal that clouds impacted most of the filters without control, for example by shutting off the pumps, though one understands that clouds are likely pervasive there. This is interpreted as INPs being captured into cloud droplets, and this is supported, but not fully clear. I was struck in Figure 6 by the fact that the CVAO INP

concentrations in 6b all appear higher during cloudy periods in comparison to the couple of periods in Fig. 6a with fewer clouds. Any ideas on why this is so?

There is only a small number of samples. It is seen that during marine events, $N_{INP}$ is lower than during the other times, even up to high ice nucleation temperatures. It is also seen that the cloud free times occurred during the marine times. However, we did not find a good correlation between coarse mode particle number concentration and $N_{INP}$. These are interesting observations, indeed. But as we only have this very low number of samples collected during the marine period, we not expand on this topic in the text.

The elevation of INP concentrations is over an order of magnitude in a few cases with the largest humps. This is unusual, but also makes me ask if the conclusions about capture of INP into cloud droplets is the full reason for differences below and within cloud. One factor could be drizzle and precipitation. It should be mentioned are solid if clouds were clearly not drizzling, as this could remove or redistribute INPs.

In the companion paper, we characterized the cloud events at MV by comparing the PNSDs at CVAO and MV. The cloud events are pervasive at MV. During the cloud events, most accumulation mode and partly also Aitken mode particles were activated to cloud droplets. We did not have a functioning APS at MV, therefore we cannot compare the coarse mode particles. However, we can assume that most coarse mode particles were activated to cloud droplets because larger particle are easier to activate. INPs are mainly in the super-micron size range, which can explain why most INPs were captured by cloud. There was no precipitation observed at the foothill, which means cloud droplets instead of drizzling were present on the mountaintop.

16) Results, page 14, line 1: Do you discuss cloud diameters in that companion paper? If not, is it consistent with some inference to cloud droplet distributions? This seems to require statement at this point, not later only on page 16.

We did not discuss cloud droplet diameters in the companion paper. But we now added the following in page 15 lines 4-6:

“These observations are consistent with results by Siebert and Shaw (2017) who observed broad cloud droplet size distributions in a size range from ~ 5 to 25 µm in shallow cumulus clouds, with the maximum of the distribution still being below 10 µm.”

17) Results, page 16, lines 21-24: It is not stated explicitly, but it seems clear that for this study, the clouds tended toward relatively high water content for marine Cu, with the lowest values equivalent to the assumptions of Petters and Wright, and the highest values exceeding Rangno and Hobbs. This is just a comment. Drop sizing or LWC measurement would be quite useful in any future studies of this type.

In this study, the LWC was calculated from Equation 6. In this function, we assumed droplet size varies between 7 and 20, with a median value of 15 µm. Our calculation of the LWC is indeed sensitive to the droplet size. However, with the assumption of a range from 7 to 20 µm, we can safely assume to cover all possible values of the LWC. As you mentioned, measuring droplet size or LWC would be very helpful in these kinds of study.

18) No reply needed, just to note that the agreement shown in Fig. 8 is striking, even stunning. Also, the discussion at the end of the Results section regarding Fig. 9 is excellent.

Thanks for saying that!

19) Discussion, page 20, lines 1-2: The referenced results for the lack of an influence of SSA on INPs is for a completely different location. I am not sure of the relevance of comparing the two studies other than to note that they concluded the same thing. Are you trying to say the SSA INPs never dominate? I would reject that notion. The only commonality in the two regions is that land sources are present at distances within a day or two trajectory distance. This is not the case everywhere.

The sentence might have been misleading, as it was not meant to say that SSA INPs never dominate. We changed it such that this should be clear now:

"This is in line with results from Si et al. (2018) and Irish et al. (2019a), both done in the Arctic, where it was also concluded that SSA only contributed little to the INP population. The commonality of these two studies from the Arctic and the present study is that land was still close enough so that terrestrial sources can have contributed to the observed INPs."

20) Discussion, page 20, lines 3-4: How does one know what maximum T desert dust is active? Doesn't this study suggests that -10C is not unreasonable?

You are correct in that there is no maximum T for the activity of desert dust particles. But we also cannot claim the opposite. As the second review suggested, samples CVAO 1596, CVAO 1641 and CVAO 1643 were heated to 95 °C for 1 hour and a large reduction of $N_{INP}$ was observed. Therefore, this paragraph was changed to:

"While the above arguments suggest that INPs in our study were mostly mineral dust particles, there were also some measurements with comparably high INP concentrations at temperatures of −10 °C and above. Although it cannot be ruled out that desert dust particles might be ice active at such high temperatures, by examining the reaction of some highly ice active samples to heating, described in Sec. 3.2.1, we found that the most highly ice active INPs on these samples were biological particles. It is an open question where these biological INPs originated. The times during which these highly ice active INPs were observed were times when air masses came from Southern Europe, travelling along the African coast and meanwhile crossing over the region of the Canary Islands. Therefore, for these specific samples, a contribution of INPs from these land sources might be assumed. "

21) Discussion, page 21, lines 5-6: Is this too prominent a question? Perhaps, but perhaps not. In regions where marine and dust populations strongly intersect, and both populations contribute to the surface area, it seems that it will ultimately be necessary to parse out the contributions. This was not done in DeMott et al. (2016), and that probably makes inclusion of those data as purely SSA somewhat suspect for the data collected in the Caribbean, especially. It makes it difficult to discern anywhere, if a few percent of dust by number is sometimes present (a few papers on this are in press). Do you have any compositional inferences to use here? Consider figures 4 and 5 for

of Gong et al. (2019a) for varied compositions during different times. Do the numbers roughly work out if you assume something to use as pure dust? You do not really dig into this at all. It may be a major question, but you appear to have some additional information that would allow you to state if your data are consistent with the proportions of mineral and marine particles.

Thanks for your comment. It is really interesting for the future work to parse out the SSA and dust contribution to INPs at Cape Verde. Since we have classified the particle types in the companion paper, we can compare the $n_s$ during marine and dust periods. We added the following in page 22, line 3:

"CVAO is a place where marine and dust particles strongly intersect, and both particle types contribute to the surface area. In the companion paper, we have classified the aerosol at CVAO into four different types. Here, in addition to looking at average values as presented above, we selected the most clean marine (CVAO 1585) and most dusty (CVAO 1591) samples for a separate calculation of ns and added the results to Fig. 9. The $n_s$ is clearly higher for the sample collected during the dusty period than during the marine period at higher temperatures (roughly >−16 °C). However, at temperatures below −18 °C it is the other way around. In general, results for these vastly different cases are both still close to the upper limit of the parameterizations reported for SSA.

These comparisons to literature raise the question if and how $n_s$ should be used to parameterize atmospheric INP measurements, which, however, is a question far too prominent to be answered in this study. In general, it is still an open issue to which extent $N_{INP}$ can be parameterized, based on one or a few parameters, to reliably describe $N_{INP}$ for different locations around the globe. It might prove necessary to develop separate parameterizations for different locations or air masses, as it was already started for parameterizations based on particle number concentrations (see e.g., DeMott et al. (2010); Tobo et al. (2013); DeMott et al. (2015))."

22) Summary and Conclusions, page 21, lines 19-20: The sentence is not complete, and it is unclear if it is referring to other studies or this one. If referring to this study, I suggest to say that "biological particles appeared to contribute: : :" I note though that no confirmatory tests were performed to ascertain biological INP influence.

You were right about the confirmatory tests, which now were added, as said before: Samples CVAO 1596, CVAO 1641 and CVAO 1643 were heated to 95 °C for 1 hour and a great reduction of $N_{INP}$ was observed. This sentence was changed to:

“These elevated values disappeared after heating the samples at 95 °C for 1 hour. Therefore, biological particles appear to contribute to INPs at these moderate supercooling temperatures.”

**Reference:**

Bigg, E. K.: Ice Nucleus Concentrations in Remote Areas, Journal of the Atmospheric Sciences, 30, 1153-1157, doi:10.1175/1520-0469(1973)030<1153:INCIRA>2.0.CO;2, 1973.

Siebert, H., and Shaw, R. A.: Supersaturation Fluctuations during the Early Stage of Cumulus Formation, Journal of the Atmospheric Sciences, 74, 975-988, 10.1175/jas-d-16-0115.1, 2017.