Anonymous review of „The ice-nucleating activity of African mineral dust in the Caribbean boundary layer“ by Harrison et al. (2022)

The manuscript by Harrison et al. (2020) presents field INP (ice-nucleating particle) measurements from Barbados in the Caribbean during a campaign of one month in summer 2017. Barbados lies within the Marine Boundary Layer (MBL), but as the sampling location at Ragged Point is elevated, direct influence of sea salt is reduced. On the other hand, mineral dust is frequently transported over long distances from northern Africa and the Sahara to the Caribbean (especially in summer) and heavily influences the local aerosol composition and concentration, and therefore the ice nucleation activity. The authors confirm a rather constant presence of mineral dust during the campaign by their APS and SMPS measurements, as well as scanning electron microscopy (SEM) and powder X-ray diffraction analysis (XRD). However, the observed INP concentration and n_s active site density are on the lower side, which is a little counterintuitive to what is expected at first glance. The authors list the observed low K-Feldspar content (0.1 to 2% of mineral dust mass) as the main reason for this comparably low INP activity, as K-Feldspar is known to be the driver of the INP activity of mineral dust. They then argue in detail why this lower activity was observed, namely a) the likely source region in West Africa having a lower Feldspar concentration, b) distinctly different transport patterns for the MBL and the Saharan Air Layer (SAL), c) physical and d) chemical processes during the transport, e) and the influence of the mixing of sea salt. These findings suggest that we should be more careful when applying INP parametrizations that are based on specific mineralogical compositions and size distributions and that a better understanding is needed for the K-Feldspar content around the world as well as the specific transport mechanisms and pathways.

Overall, the manuscript is well-written and follows a good structure. The presented analysis and figures are generally of good quality (except for some points which are listed below). I enjoyed reading the manuscript and I think it is definitely very relevant for the ice nucleation community, and fits the scope of the journal. In summary, I recommend to publish this interesting manuscript in ACP after minor revisions and after the following questions / comments have been addressed:

General comment:

The low K-Feldspar content is shown to be the most likely reason for the disparity between the observed n_s activity and parametrizations (and I agree). One of the main reasons for the low K-Feldspar contribution is then given by the transport route patterns and the potential source region of the dust in West Africa (Sahel in Mauritania and Mali). The transport of mineral dust to the Caribbean and its key features are even specifically introduced in detail in section 2. However, I wonder why the actually back trajectory analysis is then cut very short in the manuscript. From the way the arguments are presented, I expected a robust trajectory and source region analysis to confirm these hypotheses. Instead the individual back trajectories are only shown in the supplement and it is only stated in one sentence that they generally support the presented idea. I think a detailed analysis would strengthen the manuscript and may drive the point home completely.

Specific comments:

1. Fig. 1: Maybe make the background color green for the whole “Caribbean”? I think it looks a little weird as it is, or is this somehow intended?
2. P5 L138: Is the name “IcePod” an abbreviation?

3. P8 L238: It is unclear to me which size distributions (SMPS+APS or SEM) were finally used to calculate n_s. Please specify. Edit: Reading the results, I now see that the APS and SMPS were used, but it is probably best to give this information in the method section as well.

4. Fig. 3: I find it rather unusual to add a legend as a separate panel to a figure. I would suggest to add the legend below the panels a-d in a line-like format or to the right of these panels.

   Further, the numbers on each size bin composition bar are very difficult to read. Maybe you can also increase the width of the figure for better readability?

   Also on the second to last sentence of the caption there are two .. at the end.

   Moreover, as the last sentence is more interpretation of the results (however obvious it may be) than a description of what is seen in the figure, I would probably leave it out of the caption.

5. P9 L276: From Fig. 5 it seems that this concentration of 100 µg m^{-3} was not measured by the APS for some reason, but only by another method that is not described here but in another paper. I think this should probably be mentioned here. Especially, since the GLOMAP model line in Fig. 5 does not seem to see this peak in aerosol mass. Accordingly, it is maybe even unnecessary (?) trying to explain in detail why no INP concentration increase was observed when the aerosol data is partly missing.

6. P9 L278: On what observation is this statement about the excepted shift of 2°C per factor of 5 in surface area based? (Fig. 7? Literature?)

   Also where does this factor of 5 in surface area come from? At this point aerosol mass concentrations were discussed only.

7. Fig. 4: Shouldn't the line about the back trajectories and Fig. S6 be in the main text instead of the caption of a figure? I think the back trajectories are a key element for the story of the manuscript (see general comment).

   Panel c: Black, white and grey symbols are not indicated in the legend (although it is clear what they represent from looking at panel b).

8. Fig. 5a: It is unclear to me why this unusual INP "concentration" time series of the temperature at which the concentration was 0.01 INP L^{-1} was chosen. Is this somehow considered a critical threshold for Caribbean clouds? Also looking at Fig. 4a 0.01 L^{-1} seems to be a concentration at which all data points are close to or at the background concentration, if I understand the open symbols correctly. Why not choose a higher concentration (i.e., 0.1 L^{-1}) to circumvent this issue? Why not simply show the concentration at a given temperature (e.g., -20 °C, etc.)?

   Furthermore, I am having trouble understanding the green symbols: I guess the length of the vertical bar indicates the sampling time, correct? Further, sometimes there are up to three data points at one specific time, is this correct? It might be worthwhile to intercompare these samples in some more detail? It seems from this figure that there we some differences up to ~3 °C for this depicted concentration of 0.01 L^{-1}. How does this translate in variability of the INP concentration at one specific temperature?

9. Fig. 6: Some data points seem to indicate an increase in INP concentration after the heat treatment. Do you have any ideas what may have caused this? I wonder if this is a real effect caused by contamination or something else, or just variability in the measurements. If the latter were true then the language may need to be toned down a little when explaining the

differences in the text for the samples when the heat treatment did decrease the INP activity. (Although the explanations are plausible.)

Furthermore, what do the triangles mean in the figure? (Is it just to indicate the heat labile sample? If so, I wonder why the other mentioned sample is not also shown as a triangle.)

10. P10 L301 / Fig. 4c: Welti et al. (2018) present their data in detailed form for temperatures down to -16°C. Looking at the frequency distribution of measured INP concentrations at -16 °C (Fig. 5 of Welti et al., 2018) it can be seen that concentrations were usually lower than 0.1 L^{-1}. Considering this, to me it seems that extending the data from Welti et al. (2018) would result in a reasonably well comparable concentration range to what was measured in this manuscript. Maybe the generally higher concentrations from Price et al. (2018) are rather caused by the different altitude and possibly higher dust load, different dust composition or different particle size distribution at that height resulting from different transport patterns (or simply temporal variability)?

11. P12 L362: I think this paragraph is really relevant to explain the observations. I agree to the presented reasoning. However, I wonder if these arguments could be strengthened by providing a figure in the manuscript itself. For example, maybe showing a back trajectory frequency distribution from this campaign (or for summer months in general) superimposed on a mineralogy map (as described in the text) to really illustrate the point. Generally, I wondered why the information from the back trajectories were not featured more prominently here (there isn't even a mention of the Fig. S6 here), as they would be a key indicator that the hypothesis for the lower INP mineral dust activity were sound. Also maybe you can elaborate some more why you have low confidence in the back trajectories?

12. P14 L429 / Fig. 8: I wonder why you don't show the K-Feldspar-content "corrected" GLOMAP prediction in the figure (e.g., as an average line).

13. Fig. S4: I am not familiar with this method. Could you briefly describe it (here or in the manuscript) and explain what this figure means? What are negative counts? How do you estimate the relative mineral content? What is the grey and blue line? What is the x-axis?

14. Fig. S9: Is this mass fraction calculated for the surface or a mean column density?

Technical comments:

15. References should be put in parentheses ( ) instead of square brackets [ ] according to the journal guidelines

16. P3 L74: Missing space between "sources. In"

17. P4 L97: "rides above" – I am not sure about this phrasing. Maybe rephrase?

18. P6 L162: → section **3**.3

19. P6 L176: → M**ö**hler et al (2008)

20. P7 L191: what do you mean by "where the square brackets indicate the concentration"? Also there is a right parenthesis but no left one.

21. P9 L277 / Fig. 5: There is no explicitly labeled panel a) and b) in Fig. 5 yet.

22. P11 L331: SAL layer → SAL has the word layer already in it

23. P11 L337: The title of chapter 6 is rather long. Maybe it can be rephrased to a more concise title?
24. P14 L409: → Vergara-Temprado et al. (2017)
25. P14 L426: → Fig. S9
26. Fig. S5: The error bars reach out of the figure. Also, there is no mention of Fig. S5 in the manuscript yet.
27. Fig. S6: There is a weird line next to the caption and red underscore symbols after “2000” and “4000” in the caption.
28. Fig. S7: Panels e and f are already in the manuscript and do not need to be shown here again. The panels are not correctly assigned in the caption.

References:

Price, H. C., et al. (2018), Atmospheric Ice-Nucleating Particles in the Dusty Tropical Atlantic, J. Geophys. Res., 123(4), 2175-2193, doi:10.1002/2017JD027560.

Welti, A., K. Müller, Z. L. Fleming, and F. Stratmann (2018), Concentration and variability of ice nuclei in the subtropical maritime boundary layer, Atmos. Chem. Phys., 18(8), 5307-5320, doi:10.5194/acp-18-5307-2018.