Review of Hawker et al. (2020): **The nature of ice-nucleating particles affects the radiative properties of tropical convective cloud systems, submitted to Atmospheric Chemistry and Physics.**

General comments:

The authors present a very interesting study on the effect of different INP parametrizations on the radiative budget in Tropical Atlantic deep convective cloud fields. In particular, they show that INP parametrizations with a larger temperature-dependency lead to a larger increase in domain-mean daytime top of atmosphere outgoing radiation. In contrast to previous work by other authors, they present data that indicates primary ice production to be relevant also in presence of secondary ice formation. It should be clarified that the INP parameterizations tested against the Hallet-Mossop process explicitly, since this is the only SIP considered in the model.

The writing (from an editorial standpoint) is to be commended. The methodology appears stringent and valid with one minor aspect to be clarified. The work addresses relevant scientific atmospheric questions with impacts on global climate simulations. The title speaks of the nature of INPs, but the paper never refers back what “the nature” explicitly is. The topic of the paper is well suited for ACP. I recommend the manuscript for publication if the above and following comments below are addressed:

Specific comments:

Line 19-20: The paper presented tests the effect of 5 parameterizations on radiation and SIPs but the word parameterization doesn’t shown up even once in the abstract. Also, the work is not explicitly testing the impact of the nature of the INPs (i.e. bio and physico-chemical properties of INPs). It is testing the impact of the various parameterizations which are based on desert dust, feldspar and continental aerosol, so there isn’t an explicit testing of physico-chemical properties of INP on the radiation. The C86 and M92 are not aerosol based either. I suggest being more explicit about what this work does. Same goes with the title, the nature of the INP is not tested as much as the type of parameterization.

Line 22: should add “due to the Hallet-Mossop process” after “....secondary ice production” since the paper tests the parameterizations against one secondary ice process, not all.

Line 51: Add Peckhaus et al. (2016) to the references who also studied different types of feldspars and reported n_s.

line 53: Are e.g. mineral dust events not driven by meteorological factors (gust fronts, lack of precipitation, convective instability, trade winds, etc.) and have a major impact on INP concentrations?

Line 59-60: Not sure what the authors means here. Can this sentence be elaborated upon, or restructured?

Page 6 (line 165). Maybe it would help the reader to also convert the smallest allowable size of ice to an idealized diameter of a sphere, e.g. (i.e. 10^{-18} kg or ~0.1 µm in diameter)

Line 183: What errors would you expect from SOCRATES not responding to changes in ice crystal or snow number concentration, or any changes to rain or graupel species? An over- or underestimation in radiative processes? Also, how does the model then account for changes in in ICNC due to heterogeneous freezing?

Line 189: If LW radiation is only calculated for daytime, how biased is this estimate, since LW would be most effective (trapping outgoing radiation) at night time, so how will this bias the results presented for outgoing LW radiation.

Line 196: should "(ΔRad_{REFL})" come after the word "…difference" in the sentence? It seems to appear too early in the sentence.

Line 197: What do s and r stand for? It is hard to follow the equations below without knowing some sort of physical definition of s and r.

Line 201: I am not sure if I followed equation 2 correctly, but could Δcf in this equation be replaced with ($Rad_{s,cl}$ – $Rad_{s,cs}$) since you are looking at the contributions of cloud fraction to radiation between simulation s and r? If this is correctly understood, I would replace the Δcf with ($Rad_{s,cl}$ – $Rad_{s,cs}$), to make it easier to follow equation 2. Or define Δcf more explicitly than has been done in line 203.

Line 239: If the evolution of the clouds are not being discussed further, then no need to show the plots in figure 2e, f,g and h. The most useful plots are a-d since they compare the satellite with the model. Since there are no comparisons to the satellite for the other times, it doesn't give much of a validation. Also to show that the model produces a complex realistic cloud field can be demonstrated with Fig2c and 2d, so that c-h are not necessary.

Line 242: Same comment as above with Fig A2, panel c, d, e and f are not necessary.

Line 255: "flow" should be "flown"

Line 272: In the noINP case, can ice crystals that formed via homogeneous freezing, fall to lower levels and initiate secondary ice processes?

Lines 281 to 283: similar to comment above, the comparison of noINP to simulations with INP parameterization demonstrates an enhancement in outgoing radiation for D10 and A13. Can the authors clarify here that noINP simulation excludes any contribution of SIPs that could result from ice settling from higher altitudes to warmer regions of the clouds? Or is such a contribution included in this assessment?

To me this seems possible to diagnose in the model, if the assessment of precipitation evaporating results in higher humidity such that the LWP due to increase cloud droplet number. Then why isn't the possibility of ice crystals or snow settling through the clouds at Hallet-Mossop relevant temperatures allowed to produce secondary ice?

Line 285-286: "Radiative changes from the NoINP simulation to the inclusion of INP are caused mainly by .."

It is not cleat to me what is meant by this statement. I think it means the difference in radiation between the NoINP and the INP simulations mostly arises from the changes in outgoing SW radiation, but if that is the case why not state it more simply, i.e. the difference in radiation between NoINP and INP simulations …

Lines 291 – 292: The reported comparatively small change in TOA radiation when SIP is active relative to when it is inactive mentioned here, could this be because in the homogeneous freezing run SIP is by default assumed to be absent (i.e. settling of crystals to warmer Hallet-Mossop regions for secondary ice is not represented in the model)?

Line 293: The relationship between the slope of the parameterization and the outgoing radiation is implicit and not explicit. So stating that it is the key determinant seems very strong here, without clarifying that the slope is representative of the INP concentration as a function of T (Fig 1). It is the T at which a certain proportion of INPs are active is they key determinant and the physical reason for the strong influence on available supercooled liquid to be transported to higher altitudes of the MPC region. Could this relationship be clarified to invoke the temperature dependence rather than just stating the slope of the parameterization is the key determinant?

Line 303-304: This sounds counterintuitive to me because compared to the NoINP simulation, the increase in INP should increase the ICNC and decrease the cloud lifetime or outgoing SW (reflectivity) compared to otherwise what would be a more reflective liquid cloud with less ice. However further down the authors do explain why they observe this, because the liquid water path increases in the warmer part of the cloud which increases the outgoing SW. However, I find this assessment to be biased, without accounting for potential SIP in the NoINP simulation due to settling ice crystals.

Line 309/Figure 4b: I would have expected the water path due to snow to decrease because snow should leave the cloud faster than ice crystals? So why is the water path due to ice crystals decreasing? Or does it have to do with the categorisation of when a hydrometeor is considered a snow flake vs. an ice crystal in the model? Also, I would have expected that the increase in water path should be the lowest for the M92 and not for the D10, since the M92 has the shallowest slope?

Lines 313-320: If the precipitation is increased, then the snow and graupel should not be part of the cloud anymore and thus not contribute to the increased reflected shortwave radiation. If the increased condensate is falling as precipitation such that it is resulting in an increased humidity thus increasing the LWP from an increase in cloud droplets, how can it also contribute to increasing the outgoing shortwave in the cloud? It should either be classified as increased precipitation below cloud or increased condensate in-cloud leading to outgoing shortwave.

Lines 321 – 334: This explanation makes more sense and sounds stronger and more convincing to me than the explanation in lines 308 to 320. Perhaps it would be better to shift the order of these paragraphs and explain the higher outgoing shortwave by the increased CDNC at lower altitudes due to increased LWP from lower freezing rates arising from steeper INP parameterizations (which imply very little het. freezing at small supercooling).

Line 331: clarify statement more, I suggest (italics is suggested part) something like "…due to lower rates of heterogeneous freezing *at the mid-bottom region of the mixed-phase cloud (lower supercooling, Fig. 1)* and SIP at …"

Line 339: clarify by inserting “cloud fraction due to” i.e. sentence should read

“…offset somewhat by decreases in the cloud fraction due to homogeneous freezing in the ~10 – 14 km regime (Fig 6a)”

Line 358-360: If this is true, (and it sounds like a good explanation), shouldn’t the decrease in outgoing LW shown in Figure 4a be the highest for A13 and not for C86. Because A13 results in the highest number of ICNC at the top of the MPC region and in the homogeneous freezing region therefore should trap most of the outgoing LW radiation thus giving the most decrease in the outgoing LW.

Line 379:. Change to “It has been argued that the observed (or derived) primary ice particle production rate…”. Otherwise, the statement is false, because if the primary production rate is high, the secondary ice production (H-M process) would be low but still present, primary ice production would in fact dominate cloud properties.

Line 384/ line 219/lines 440-445: Most relevant comment. You show that higher primary ice production rates in the temperature range between 253 and 238 K, e.g. in A13, have a large impact on the total on top of atmosphere outgoing radiation, yet you exclude SIP which are active at colder temperatures than 265 K. Can you elaborate on the expected impact/uncertainty in your results and concluding statement, (SIP is less important than primary ice production) stemming from your simplification that SIP is only including Hallett Mossop process? Please justify why your concluding statement is valid.

Line 383-384: This is an important outcome of the study, but should be caveated with the notion that other possible known and unknown SIPs are not considered. The authors in part do that by acknowledging in parenthesis that the SIP considered is the H-M, but I think they could go one step further in saying that this could change with more parameterizations and quantification becoming available for lower temperatures where SIP becomes important say below 265 K (Lauber et al., 2018).

Page 14 (line 385). An average impact comparison in the text might be supportive for the reader (e.g. mean over all parameterizations total INP impact 9.8 W/m2 to total SIP impact 2.7 W/m2)

Line 453-455: A possible explanation for this statement should be given here in in the conclusions, since this is an important point or outcome of the study. Have the other studies that are mentioned in these lines also only tested the influence of the Hallet-Mossop process? If not, this should be clarified. Further, since this has evaluated the influence of SIP due to the HM process, it should be stated here in the conclusions.

So this conclusion is true, when the SIP being considered is HM. But it remains open if the conclusions would still hold if freeze shattering and other mechanisms (e.g., as described in Lauber et al., 2018) were included in the models.

Line 496: In addition to Holden et al. (2019) the authors could add Coluzza et al. (2017) and Kanji et al. (2017) since that lack of knowledge on what constitutes the identity of an active site was already discussed in these publications.

Line 497: The last statement here has surely been mentioned before by other authors in numerous publications. While it is valuable that the authors also come to this conclusion (need for INP measurement across the entirety of the MPC regime), this study is not the first to recommend such an outlook and the sentence can be modified to say "..as reported before by XX.."

Figure 2: Should there not be a "radiation" in the color bar caption, e.g. Outgoing OA longwave radiation (W m^{-2})?

Figure 5: Colour legend is missing. I suggest adding it here even if it is the same as previous figures for ease of reading.

Appendix A, page 34 (Figure A1). The three kind of blue lines are not easy to distinguish from the black line.

Appendix A, page 34 (Figure A1). What does the unit of / 10^8 m^{-3} mean? Is it two particles per 10^8 m^{-3} of volume?

Figure A3 panel a: what are the regions filled with black colour? Could the colors be changed so that the contrast between green and blue is better visible? If black is just the border of the bars, I suggest removing the borders since it reduces clarity of the plot suggesting that there is a third colour.

Line 810: Histograms should be singular not plural.

References

Coluzza, I., Creamean, J., Rossi, J. M., Wex, H., Alpert, A. P., Bianco, V., Boose, Y., Dellago, C., Felgitsch, L., Fröhlich-Nowoisky, J., Herrmann, H., Jungblut, S., Kanji, A. Z., Menzl, G., Moffett, B., Moritz, C., Mutzel, A., Pöschl, U., Schauperl, M., Scheel, J., Stopelli, E., Stratmann, F., Grothe, H., and Schmale, G. D.: Perspectives on the Future of Ice Nucleation Research: Research Needs and Unanswered Questions Identified from Two International Workshops, 8, doi:10.3390/atmos8080138, 2017.

Holden, M. A., Whale, T. F., Tarn, M. D., O'Sullivan, D., Walshaw, R. D., Murray, B. J., Meldrum, F. C., and Christenson, H. K.: High-speed imaging of ice nucleation in water proves the existence of active sites, 5, eaav4316, doi:10.1126/sciadv.aav4316, 2019.

Kanji, Z. A., Ladino, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D. J., and Krämer, M.: Overview of Ice Nucleating Particles, in: Ice Formation and Evolution in Clouds and Precipitation: Measurement and Modeling Challenges, 2017.

Lauber, A., Kiselev, A., Pander, T., Handmann, P., and Leisner, T.: Secondary Ice Formation during Freezing of Levitated Droplets, 75, 2815-2826, doi:10.1175/JAS-D-18-0052.1, 2018.

Peckhaus, A., Kiselev, A., Hiron, T., Ebert, M., and Leisner, T.: A comparative study of K-rich and Na/Ca-rich feldspar ice-nucleating particles in a nanoliter droplet freezing assay, Atmos. Chem. Phys., 16, 11477-11496, doi:10.5194/acp-16-11477-2016, 2016.