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Comment on acp-2021-927

Anonymous Referee #1

Referee comment on "The influence of multiple groups of biological ice nucleating particles on microphysical properties of mixed-phase clouds observed during MC3E" by Sachin Patade et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-927-RC1, 2022

The manuscript, "The influence of multiple groups of biological ice nucleating particles on microphysical properties of mixed-phase clouds observed during MC3E " by Patade, et al., explores the impact of primary biological aerosol particles (PBAP) acting as ice nucleating particles (INPs) on the properties of the microphysical processes (including, ice formation, cloud droplet, precipitation) of the mixed-phase clouds. They used for that a mesoscale model called AC and they implemented an empirical parametrization for ice nucleation that can distinguish between the different types of PBAP and has been derived based on data collected during a measurement campaign in the Amazonian area. They simulated a mixed-phase case study in the USA during MC3E campaign and they compared the simulated output with these observations. They ran several sensitivity studies where the initial concentrations of PBAP are changed or some SIP mechanisms were turned on and off to investigate the impact on the cloud and precipitation formation.

In general, this topic is important. Moreover, these types of modelling studies are interesting and potentially quite useful for the atmospheric and aerosol-climate community who wish to model heterogeneous ice nucleation by PBAP INPs and evaluate their competition with secondary ice production to eventually reduce the large uncertainty in aerosol-cloud interactions processes. However, with my full review of the manuscript, I have several major comments, questions, and suggestions that I feel the authors need to address before I recommend the manuscript for final publication in ACP.

General comments/questions:

1- The PT21 used here was derived from real measurements in the Amazonian area (mainly forest type of land surface, maybe mixed with some wetlands). In lines 366- 369, it was stated literally that “PT21's observations were used to calculate the relative contribution of various PBAP groups to insoluble organics. The parameters for the shape of PSD of each PBAP group (modal mean diameters, standard deviation ratios, and relative numbers in various modes) are prescribed based on observations from Amazon (PT21)". Having that said, how can the authors justify the use of such a parameterization on a different type of land surface/case study, where the aerosol load are/might be different and accordingly their prescribed parameters for the shape of PSD of each PBAP group could also be different? Can the authors recommend using such a parametrization in global models as well to represent also the PBAP from different types of land surface and marine areas? If yes, are there any requirements or limitations that one needs to take into account before using/implementing it?

2- Throughout the whole manuscript and when discussing the results, it was difficult to follow up on the calculated percentage values of the changes of some prognostics and diagnostics that were shown in the figures, but as vertical profiles. Therefore, it can be great if the authors provide at the end of the manuscript a table that summarizes the vertically summed domain averaged values of the model diagnostics (including, LWC, ice concentration, precipitation (convective, stratiform, and total), short and long-wave radiation/flux, and cloud fraction) for the different simulations and compare them with observation if relevant (or possible) or with other studies that were mentioned in the text even for a different region-domains/case studies. Such a table can provide an overall overview of the whole story of this manuscript that can support the conclusion of PBAP relevance to ice formation, make it easy for the reader to estimate the changes and their corresponding percentages from one simulation/scenario to another as well as improve the overall quality of this manuscript.

The author can choose to provide one table or two tables (one where they vertically sum the values from the surface to the level of -35 C, where heterogeneous freezing is relevant and another one for full vertical summed values including the homogeneous freezing).

3- Due to the high number of simulations, it was not clear if some of the ice processes were turned on or off in each simulation especially since it was sometimes confusing which one is turned on or off throughout the text (Fx, Line 782 - 783, where the authors suddenly mentioned that the SIP was turned off in the control simulation after the impression that it was on from the beginning). Therefore, another important table is necessary for this manuscript that shows the different configurations of each simulation in this manuscript. Fx, the table summarizes the 1- different types of PBAP or other INPs that were considered in each simulation, 2- simulation names, 3- simulation configurations, 4- the cloud ice process which was turned on/off, 5- the initial and the increased concentrations of different INPs considered for each simulation 6- as well as the corresponding figures numbers that are resulted from each simulation.

4- Vertical profiles are important, but spatial distribution (i.e maps) of the diagnostics (vertically summed or averaged) to show the spatial variability for the different simulation/scenarios could improve the quality of the manuscript

5- The manuscript does not provide any supplementary material that could be useful (I have added further some suggestion on how could it be)

6- Modelling uncertainties and the term "good agreement": This manuscript always addressed the uncertainties of other work/observations and never mentioned the uncertainties that could result from running any type of atmospheric model including AC (it's not an exception). This in return led the authors to state "good agreement in many places throughout the manuscript", where I would be careful to say "good agreement" but rather use "acceptable agreement", given that both models and observations have/should have uncertainties. Since the authors addressed one of them, then it's fair to estimate and address the model range of uncertainty. This is important to avoid giving the readers the impression that only observations have uncertainty and model never have that and they are always perfect, although it's not mentioned explicitly.

Major comments/questions in detail:

1- Lines 199-200 with Figure 1: Where the value 2355 J kg^{-1} can be read from the figure? And why it's relevant to be mentioned here? The description of Figure 1, b in those lines does not correspond to what one can get from this part of the figure?

2- Lines 201-202 with Figure 1: Same as the previous comment. At this level (840 hPa) neither the air temperature nor the dew point temperature was 14 C as stated in the text. Which line in Figure 1, b shows that the temperature at that LCL (840 hPa) was 14?

3- Please show the right figure that corresponds to the text or change the text according to what can the reader sees from the figure.

4- Lines 202-203: What is the estimated amount of vapor in the entire depth of the troposphere? It seems that this sentence is not complete and this piece of info is missing in the text. Either remove the whole sentence if it's not used further on in the text or type the right value and consider fixing the sentence.

5- Lines 199-203: This part needs to be revised/rewritten.

6- In figure 1, the resolution of figure 1, b should be improved. It was hard to read and extract information from it with its current resolution and check the values stated in the text. Please consider providing a better version with a higher resolution/readable Skew- T plot.

7- Figure1, c: Would it be possible to provide the uncertainty in the modeled line as well similar to the observation as the predicted/simulated CCN concentration is deviated from observation? Add some text as well in the manuscript to describe this part of the Figure.

8- Lines 271-282: I would guess that those aerosol measurements from IMPROVE were not on the same dates as the MC3E campaign as they seem to be a separate data-set. But can the authors give more info about those measurements (dates, some time series of the measured species especially for PBAP), and more importantly the scaled profiles of aerosol mass concentration that matched actual measurements, which were mentioned in lines 280-281? Those can be added to the supplementary material and referenced here in the manuscript.

9- Lines 307-310: How those processes are different in terms of temperature? It's better to add (in parentheses) the range of T where each of those SIP mechanisms is more efficient/relevant.

10- Line 344: As the parametrization PT21 has been used in this study, I suggest adding & present the formulation of PT21 with its used/corresponding parameters here in this manuscript at the end of this section 3.2.

11- Line 347: Where those initial and evolving boundary data for meteorological conditions were taken from? Did the authors use any other climate/regional model to derive them? Or they were taken from observations? If any, this should be mentioned more clearly. Better show some meteorological conditions plots for the simulations in the supplementary material.

12- Line 354: what was the spin-up time for the conducted simulations in this study?

13- Line 371: It would be a good idea to plot an example of those aerosol initial/prescribed profiles together with the predicted aerosol size distribution from AC, especially for PBAP (can be added to the supplementary).

14- Line 374: Now I see the text that explains Figure 1c. either move the text further up or consider moving this part of the figure and separate it from Figure1 and move it here as it can stand alone or join it with the suggested other plots from a previous comment in the supplementary (see the above-mentioned comment on Line 347). The whole of figure 1 can go to the supplementary.

15- Line 379: What was the uncertainty range of the modeled CCN concentration? Consider adding it to Figure 1c.

16- Line 381: Since the uncertainty of the simulated line from the AC model is not provided, I don't agree that they are in a "good agreement", but rather in an "acceptable agreement"

17- Line 385: was the SIP and homogeneous freezing turned on or off in the control simulation? Was there any other type of INPs activity (i.e., dust) considered in the control simulation in the new version of AC? Better mention those explicitly if they are turned on by default after adding the PT21.

18- Line 385: For validation reasons, why the authors did not compare their PT21 with another parametrization for this case study, fx, the older version of PT21?

19- Line 397: What does TWC refer to in the Figure 2 caption?

20- Line 435-436: Figure 3a How authors can read agreement from that subfigure? does not show an adequate agreement between observation and simulated averages of LWC as the authors wrote in the text. It's clear that the simulated means are deviated by nearly one order of magnitude from the observed means in the convection case below 0 C. Maybe, one can see some sort of agreement in the stratiform case.

21- Line 438-444: same as in the previous comment. What about the points in Figure 3c at T around 15C? Here, the simulated domain averaged points deviate from the observed mean values by ~ factor of 2. Again, this difference needs to be stated clearly in the text and then evaluate overall and say that there is an overall agreement. I would rather use the term "an overall acceptable agreement" rather than "a good agreement" here. I would also suggest adding a calculation of the percentage range of that agreement (do the same for the above comment in Fig 3 a,b)

22- Lines 470-475: How the authors can justify that two-thirds of an order of magnitude bias between observation and modeling could be better than half of an order of magnitude bias?

23- Line 472: Was the underestimation of "measured" or "simulated" ice number concentrations? Because the author said "also" at the start of this sentence.

24- Line 497: from Figure 4c the bias in reflectivity between 3 – 8 km is higher than 8 dBZ, Look at the first 5-6 points. The bias at those points is at least 10 as shown in the figure. So fix that range in the text.

25- Figures with the vertical profiles: Since not only observations have uncertainty, but also modeling output especially since the points are mean values, it's a good idea to add the error bars (uncertainty) to the simulated mean values of the diagnostics that are shown in these figures similar to the observations.

26- Lines 510-511: Again, the term "good agreement" does not fit well here. Same as previous comments. Here, the authors stated clearly the 1-2 hours delayed simulated peak of the precipitation and justified that by the uncertainty of the initial and boundary conditions of the 3-D model, and then they wrote that there is a "good" agreement between the observation and the modeling results. What criteria do the authors define for "good agreement"? Please consider fixing the term "good" here and revise throughout the whole text and use it only when it's relevant.

27- Line 521: Where the total was estimated? was the 0.3 L^{-1} calculated by the model or detected by other observation or modeling studies? If it's calculated by AC, then add the total to figure 5. If it's from other studies, then consider adding a reference?

28- Section 4.3 and figure 5: How the fraction of the number concentration of PBAP INPs was calculated from the scaled profiles of mass mixing ratio that were used as input fed into the model? As the authors mentioned earlier in lines 271-280, they used some observations to estimate those aerosol profiles (Table 2). But they never mentioned or explained how the model calculated the fraction of PBAP INPs from the input mass mixing ratio? how was the size distribution of the PBAP in this domain (consider providing these to the supplementary)? Was there any other assumptions to calculate the PBAP INPs fraction (i.e., the mass of the bacterial cell, fungal spore cell, or their particle densities)

29- Figure 6-a: Was primary ice (PRIM) referred to by the first blue bar for all INPs considered in the AC model or only for PBAP?

30- Line 552: Use the same name/short name that is used in Figure 6-a for consistency. The authors used HOM for the homogeneous drop freezing and PRIM for the primary heterogeneous ice freezing. Better to name them primary Heterogeneous freezing and primary homogeneous drop freezing.

31- Lines 565-567: Make sure of the estimations of those percentages and if the authors can calculate them then it's better to show them in a subfigure versus T?

In detail, if ice-ice (blue dots and line in figure 6-b and c) contributes 50% and sublimation (green) contribute 7% as stated in the text then the rest (red for active INPs, brown and pink for the other SIP) should contribute 43% to complement the 100% for the total (black) at T = -25 C. Let's assume that each of the rest contributes similarly as sublimation (green) does (7%) although they show a much lower contribution from figure 6 b and c, then they will sum up to 21% and not 43%. Is there some other source missing in this figure that could complement 100%? Or it's just the wrong estimation of the percentages? The same goes for the stratiform case.

Therefore alternatively, I would suggest adding subfigures (6 d and e) where the authors display the percentage contributions of each process to the total ice number concentration (x-axis) vs T (y-axis)? Add the homogeneous contribution as well as figure 6-a shows the largest contribution to the total ice, especially at low T than -35 C.

32- Lines 573-575: This may create uncertainty in estimating the HM contribution to total ice. Can the author give an estimate for this uncertainty?

33- Section 5.1: Did the authors consider dust or any other type of INP than PBAP in those simulations including the control one? If yes, what was the ice nucleation parameterization for dust used in these simulations in AC?

34- Lines 615-616: Please consider revising/rewriting this sentence. Are the authors comparing convective to stratiform here? Less than 50 % of what? In which case, stratiform or concoctive or both?

35- Lines 644-646: Interesting and maybe unexpected result here, however, the justification is NOT relevant here. To my knowledge, homogeneous freezing works at T lower than

~ -36 C, so it should be insensitive and inactive at -20 C. Only Heterogeneous and SIP can contribute here at this level (range of T)? This leads to a couple of questions; how did the authors define homogeneous freezing in AC? Which parameterization is used for homogeneous freezing? What is the range of T for homogeneous freezing in AC?

36- Lines 647-650: Can you explain more clearly why this is happening?

37- In Figures 4 a and b, the AC model with its parameterizations was underestimating the observed ice concentration already. It was not clear if SIP was turned on or off in the control simulation to explain this deviation. assuming that homogeneous freezing was inactive at -20 C, can the authors clarify more properly why adding more PBAP could result in less ice concentration at levels higher than -20 C where most PBAP is relevant for ice nucleation?

38- Lines 640-650: Does AC take into account the dissolving factor/fraction of each type of aerosol considered in AC? If yes what was the assumed value for each type of PBAP?

39- Figures 4, 7, and 9: Why the ice concentration from the control run in Figures 4 a,b is different from Figures 7 d and 9 d, especially at T lower than -20 C? Should not be the same control simulation/run in 4, 7, and 9?

I suggest unifying the x-axis range in all the above-mentioned subfigures, especially for ice concentration, and making it from 10^{-3} to 10^{1} cm^{-3}, so it’s easier to compare.

Also, add the observation data points from Figures 4 a, b to Figures 7, d, and 9 d as they are also relevant here.

Something is missing/ or not explained properly in justifying these results (figure 7 and 9).

40- Line 653: Can the AC model distinguish how many ice crystals that are resulted from “downwelling” homogeneous freezing from the other amounts of ice crystals that are resulted from other processes (heterogeneous freezing and different SIPs processes) and aloft up at each level of T, especially at those higher than -20 C, where homogenous freezing should be inactive? if yes, could you please explain shortly how?

41- Figure 10: Are those diagnostics produced explicitly by AC when all SIP and homogeneous freezing were turned on? Or they were estimated from different simulations where only one process turned on and the other were off?

Why the very high bac simulation contributes to more ice budget/concentration (green in 10 a for primary ice condensation-depo cold and warm) than the control, which is contrary to the conclusion that the authors drew from Figures 7 and 9 d where they claimed that the control simulation contributed to more ice concentration?

42- Lines 684-686: Again, same comment as the previous one. It seems somehow that what is shown in Fig 10 a and explained in those lines is contradicting the conclusion drawn from figures 7 and 9 d? Any explanation? Please clarify the explanation and the justification of those results?

43- Lines 693-698: this is an important conclusion, but the plot to show that ratio is not shown in the manuscript. I suggest showing the ratio plot between SIP and primary ice without homogenous freezing.

44- Lines 715-720: I suggest adding a table where the authors summarize the vertically summed and domain averaged values of precipitation, ice concentration, and LWC for the whole period of simulation for the different simulations and scenarios. This can give a better picture of those changes stated in the text.

45- Line 730 and figure 12: These short and longwave radiations/fluxes are a result only of cloud-radiation interaction (indirect effect), right? If yes, Can AC also check the direct effect resulting from PBAP-radiation interaction? or did the authors run any simulation to check only the direct effect and how different the values would be from the indirect effect?

46- Lines 756-768 and figure 13: This makes a lot of sense, and that's what one would expect, the concentration of cloud ice will increase by increasing the bacteria or fungal concentration or both. Now, how relevant this change is, is another story that depends on many factors. However, my question is, why this was not the case in Figures 7 and 9 d when PBAP concentration was increased? Any justification? Can one conclude here that there is a competition between the different types of PBAP or INPs when they are all included in the simulation (control), so they prohibit each other efficiency in nucleating ice, whereas this competition is canceled when only one type of PBAP is considered and increased?

47- Lines 781-783: Difficult to recognize which process is turned on or off in each simulation, therefore, hard to follow or assess the rest of the text in this section.

48- In lines 782 - 783, NO!!! Are the authors still talking about the same control simulation that has been discussed earlier in the previous sections (which I think it's the case) or it's a different one? If it's the same one, why did the authors wait until here to mention that one or more SIP mechanisms were turned off in the control simulation (this should have been said much earlier)? If this is the case then, What's the point of comparing simulations with no SIP to the control simulation where SIP was turned off as well?? In this context, I would suggest to the authors add a table that summarizes the different simulations' names, types, and different configurations and processes that are turned on/off for each simulation. (See my general comment)

49- Liners 791-793: Since the authors mentioned this conclusion, why the figure is not shown here to support the text? Consider either adding the subfigure or removing the text.

50- Lines 795 until the end of this section: I'm lost, which simulation refers to the inclusion of SIP that the authors are talking about here in the text and Figure 14? Is it the control (that's what I can hardly guess from the legend Figure 14), but this again contradicts what has been said and assumed by authors in lines 782-783?

51- Line 807: warmer than -25 C as stated in the text or -15 as shown in figure 14 d? 52- Line 809: warmer than -15 C as stated in the text or -25 as shown in figure 14 c?

53- Lines 823-825: Any justification for such a result here, since SIP is known to efficiently enhance precipitation?

54- Line 828: I think this section is important and needs some more explanation and justification as it clearly shows the importance of SIP vs PBAP, especially in the convective case at T higher than -30C. For example, why in figure 15b (stratiform), the ice concentration resulting from the control was nearly one order of magnitude higher than the other two simulations with no SIP at T ranging from 0 to ~-13 C, and the opposite happens at lower T than -13C? Adding a sentence or two at the end of this section summarizing the conclusion of this section would be good.

55- Lines 870-872: Figure 4b (stratiform) in the range of T between -10 and -16C the bias was more than a factor of 3 (at least half order of magnitude)

Consider having a closer look at the COMP Obs (pink points) and HVPS Obs (cyan points). This range of bias between simulated ice concentration and observation is almost in the same range as the change resulting from removing SIP (Figure 14, d), where authors eventually concluded that this change is large indicating the importance of SIP in ice production.

56- Yes, observations may indeed have uncertainties, but all models also have uncertainties. Since the model uncertainties were not shown, so, the AC model can also underestimate the observed ice as clearly shown in Figure 4 in both convection and stratiform.

57- Lines 876-878: See the above comment. What about the half order of magnitude difference in the ice concentration and the 1-2 deviation in capturing the precipitation peak taking into account the relatively short time of the simulations (48 hours)?

58- Lines 891-893: Yes, it might be that dust and BC concentrations are much higher than PBAP at T -15 C in the chosen domain or at any different domain (also globally), but do the authors think that dust and BC can initiate ice nucleation at this T or higher?

59- Lines 909-911: It's good to mention "In our study". Although such a sensitivity study is important in a high-resolution mesoscale model, however, this study has a few limitations that should be mentioned especially when talking about the shortwave and longwave flux radiation. Those limitations are 1- the small chosen domain representing maybe one or two types of ecosystems and not a whole globe, 2- the limitation in the vertical resolution (model top was at 16 km) and not the whole atmosphere, and 3- it's a mesoscale model and not a climate model to eventually get a global conclusion on the impact of PBAP.

Minor comments

1- Line 42-43: How little is the effect on the ice phase in the convective region? Consider providing a value similar to the stratiform region.

2- Line 46-47: Same comment as above. Provide this no significant in number or percentage?

3- Line 47-48: Same comment as above. How little is the effect on surface precipitation as well as on shortwave and longwave?

4- Lines 57-64: although those are well known, consider adding a reference at the end of each sentence in those lines. There are many.

5- Line 74: Only insoluble material in the PBAPs? Please provide a reference here? 6- Line 90: make sure of Hummel et al 2018 whether it fits here?

7- Line 102: consider adding a reference at the end of the sentence here.

8- Line 178: Be consistent, either PBAP or PBAPs (you decide) throughout the whole manuscript

9- Figure 1, remove one of the (c) on the right or left side of that part of the figure/plot (CCN Conc vs Supersaturation)

10- Line 277: Number concentration or mass concentration? Please be more specific. 11- Line 341: What does ATTO stand for?

12- Line 369: add the paper reference here.

13- Line 459: be consistent with the unit! Either use L^{-1} or cm^{-3}. If figure 4 shows it with cm^{-3}, then try to use/unify that throughout the whole manuscript.

14- Line 494: remove "illustrates" after "This".

15- Lines 520-521: similar to the comment in line

16- Figure 6-a: The ticks need to be fixed to match the corresponding bars

17- Line 772: I think the precipitation is shown in Figure 13, f and not So, consider changing that in the text.

18- Line 913: consider removing the second "affected"

19- Lines 1073-1078: The same reference is written Put the full citation of part I and remove one of the copied part II citations.