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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurement report: Ice nucleating abilities of biomass burning, African dust, and sea spray aerosol particles over the Yucatan Peninsula" *by* Fernanda Córdoba et al.

Anonymous Referee #2

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In the here presented study, atmospheric aerosol at two different locations on the Yucatan peninsula (Mexico) was examined, with a focus on Ice Nucleating Particles (INP). Three different types of air masses were examined, and besides for INP properties, general aerosol properties and results from a chemical analysis were made in addition.

The work is adding to the valuable growing body of studies on atmospheric INP from a region where previously no measurements had been made. The instrumentation used for the INP measurements is a still quite novel one. So throughout the text a feeling arose that comparisons with literature were sought, maybe to confirm the measurement

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method. However, and this is one of my main criticism, are not always useful in the way they were made (more on that below at “specific comments).

While the structure of the text is fine, it is occasionally difficult to follow when too many numbers were given in the text instead of discussing the general results. Again, more about this is given below.

My main concern is about the method used for determining INP concentrations. Likely, I assume, the issue is about the respective description of the method, which is not sufficient. And again, find more on that below at “specific comments”.

Additionally, the discrimination of the samples into three different phases was not motivated in a satisfactory way. It was not clearly described if/how samples were chosen amongst a larger batch or if they were really only discriminated through the time when they were samples. It was particularly shown that during the three different phases, there were variations in the chemical composition. That questions the discrimination into different phases as they were done. More on that also below.

Having said all that, the data and the manuscript as such are still valuable. So overall, I am sure that this manuscript can very likely be changed such that it can be published in ACP. However, major revisions are needed.

Specific comments:

Before going through the text line by line, up front here are my comments / concerns about three separate topics as indicated in the above text. I apologize for repetitions:

(1) Comparisons you make in the manuscript with data from literature do not always seem useful. While some of that is referred to again below, here my main concerns: You measure at different locations than other cited studies, with different distances to sources, so concentrations necessarily have to be different. In this respect, comparing concentrations does not make sense. Onset-temperatures are not useful for comparisons, either, as they depend on the amount of material sampled and the measurement

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method used. Surface site densities (n_s) are useful for comparison, but only if the surface area is related to the same aerosol particle "type": some studies only look at one specific type of INP as for example only a mineral dust (chamber studies) and will result in n_s for these dust particles, while other studies examine atmospheric particles and use the surface area of all particles and will then result in n_s for this type of air mass. This is not well discriminated in your study and below some specific parts of the text related to this issue are given.

(2) Concerning your method: There are orders of magnitude more particles than there are INP, as you describe quite correctly in the introduction. Particles can be so small that you will not see them with your microscope (Fig. 2b will only show the largest ones). One droplet will certainly have a multitude of particles in it (compare Fig. 2b with 2d). That alone explains why you obtain useable results by looking at a comparably small number of droplets. But then, which fraction of the surface (i.e., of the sampled particles and therewith INP) is not included in droplets (between the visible droplets, but also on parts of the glass slide that are not examined at all)? And how many droplets will have more than one INP in them? How much does that influence the results? This is certainly included in the factors that are used in equation (2), but this is so essential to your method, that you can not only refer the reader to another publication. This needs to be explained in the manuscript (main text or at least in the SI). Some specific issues regarding your method already here:

line 237-238: How stable is your temperature calibration? I somehow assumed you would use a paste that ensures good thermal conductivity between different metal parts of a set-up. Have you ever tried that?

line 266 and Fig. 2: How is the desired size decided? How about droplets containing more than one particle (which will have to be the case)? Fig. 2 looks as if droplets "eat up" other droplets. - How about one droplet including several INP?

Maybe that all is included in the corrections factors given in equation (2), but as this is

a VERY crucial issue, this needs to be discussed in here, instead of only referring to another paper.

(3) The evaluation of the data based on different air masses which are assumed to predominantly appear during certain times of the year has its weaknesses. There are some high temperature INP that are ice active above -19°C for samples classified as both MA and AD which are absent for BB. Other than that, all data are quite similar. The comparisons of data from other locations, in which more clearly only certain air masses were measured seems a bit arbitrary, particularly as the results in here are not traced back to peculiarities of different air masses. Instead, the argument is that INP concentrations for MA are higher than at other marine locations because there were cold fronts with high winds while INP concentrations for AD are lower due to the larger distance. That all might make sense, but it could also be that AD and MA are similar in concentration as the sources in both cases were the same! Also, for BB, concentrations are low - but what was the background concentration before that air mass entered the BB region in Yucatan? Instead of comparing concentrations, which necessarily become lower further away from sources, it would be beneficial if only parameters as n_s would be compared. But as said above and again also below, care has to be taken to only compare to studies which were based on the same determination of the surface area (using all particles and not only a sub-fraction).

line 49: “more than 50% of the precipitation is initiated via the ice phase”. It is not as easy as this - in the work you cite here, it is shown that this fraction depends on latitude and on land versus ocean. This number (50 %) for a global fraction is not given in the cited study, and I would advise you not to give such a clear number if it is not scientifically sound. "Large fractions" or something like that seems more appropriate.

line 64-65: “Only 1 in 10ˆ5 to 10ˆ6 of the aerosol particles can act as INP at temperatures higher than -38°C” - I know that something like that is often said, and I didn’t check the citation here, but it is difficult to give such a fixed number, at least if it is not related to a temperature. If you look at the range of INP concentrations you’ve reported

in the abstract, you'll see that this already covers almost 4 orders of magnitude. And your values do not cover the whole temperature range from -38°C to 0°C, so the total range is MUCH larger. This shows that values within (only) one order of magnitude for the whole temperature range > -38°C, as you indicate it here, do not make sense. Saying "roughly one in a million at -20°C" would be a much better way of putting it. That can be estimated based on atmospheric samples in e.g., Petters & Wright, 2015.

line 122: An even broader dataset on Arctic ambient INP was published in Wex et al. (2019), even pointing towards an annual cycle in INP concentrations across the Arctic.

line 126ff: This comment concerns all about the Welti et al. (2020)-ACPD-paper: It is a bit strange to give the results such detailed for a study that is still in discussion. Also, it is not clear why you give these values so detailed, anyway. If you want to compare with values from your own results, then give these details later. Here, in the introduction, these many numbers only make the text very difficult to read. I suggest to delete all these numbers from line 123 to line 138 and instead give only one or a few important statements from this study. Also a small remark: Is it generally know what "N-temperate zone" is? I guess this is an expression used in the here cited study, and if you want to use it, you have to define it, too.

line 143-146: DeMott et al. (2003) even showed that INP from the Sahara were found over Florida.

line 158-159: Do you know the study by Schnell & Vali (1976)? They already report a dependence of INP concentrations with climate zones, albeit only based on leaf litter - read it and then decide yourself if it fits in here.

Table 1 and related text: CPCs are mentioned in Table 1 but not in the text. If they were used for the here presented analysis, add a description to the text. If they were not used in this study, delete them from the table. Also: Which CPC was used in Merida in 2017? Please specify in the table! Also: Mini-Vol was an abbreviation used in the text, in the table it's MiniVol. Unify!

line 196 and line 369-370: This is one of my main concerns: How were the samples included in this study chosen? Randomly? Or for which other reasons? Or were all collected filters included? If not, Did the choice of a subset influence the conclusions drawn from the study? As I understand, you just attribute different air masses to different seasons? Is that justified? There is a study by Wex et al. (2016), in which within one month on Barbados different air masses were observed (clean marine, marine with new particle formation, and African Dust), all advected across the Atlantic. That was the case in November, and again in April. Similarly, a certain air mass likely cannot be expected to be connected to a certain season at your sampling locations. Therefore: It at least has to be mentioned how you defined/chose the different air masses, and how you checked if all air masses during a sampling time belonged to the type of air mass you expect during this season.

line 215-216: How much time passed between sampling and analysis? Give that number here. Have you ever tested if the INP concentration is influenced by storage at temperatures above 0°C? Most studies I am aware of freeze their samples prior to the analysis.

line 370ff: You show in Fig. 5 and the related text, that "background conditions" were different from other days during the three different air mass types (particularly pronounced, to my understanding, for AD). How was this variability treated when you then summarized the results to obtain one value representative for the whole air mass (as done for Fig. 6 and Fig. 8)? This is, again, linked to how you chose the samples that are presented here.

line 392: Of course, if there is also a marine influence (which is to be expected), then part of the sulfur will also some from the same sources as described for MA above. Reformulate this passage accordingly!

line 429: Am I right to assume that you calculated the particle concentration based on the measured size distributions? Please mention explicitly how concentrations were

obtained!

line 453-454: There are trade wind cumuli across the Atlantic and they can produce warm rain! Just for fun I opened "windy.com" while writing this, checked for clouds between Africa and Yucatan, and (as I expected and as you will always see) there sure were several locations with intense cloud fields. Please correct this statement! It is clear, though, that particles are transported in "SAL" (Saharan Air Layer), and yes, "trickling out" from that indeed happens. But wet removal likely occurs, too.

line 461-463: Was the study you cite here based on a lab-experiment? If yes, then the dilution of the air also will have played a role, and then it is not clear to me how representative this is for atmospheric concentrations, and this quote might not be very meaningful.

line 467: In Tab. S1, there are 7, 5, and 4+4=8 samples for MA, BB and AD, respectively. The number of data-sets shown in Fig. 7a seems lower. Is that so, and if yes, why?

line 471-473: “onset freezing temperatures” are not a good value for judging a data-set, as they will depend on the amount of air sampled and the instrumentation used for the evaluation. At least add this information, or better delete this sentence completely.

paragraphs starting at 475 and at 488: It is much better to give comparative data in a figure instead of giving a range of numbers in a text. Additionally, a concentration of INP always has to be given at a temperature. Giving a range of concentrations spread over a range of temperatures is not helpful, and comparing concentrations obtained at different temperatures or for different temperature ranges is not meaningful, either. I suggest that you make an additional figure with your data with separate panels for the three air masses in which you always add the literature data you want to compare to. By reading this, one cannot understand the relation, and with this new figure you can avoid text filled with numbers which is difficult to read. And, as you quite correctly say, you are far away from the source, compared to the other studies to which you

are comparing to, so that comparing a concentration is not the best thing to do, as a concentration HAS TO decrease further away from a source. Comparing intrinsic INP properties, such as n_s, makes more sense.

line 489: Actually, INP concentrations for MA and AD are surprisingly similar, both in the range they cover and in their lowest and highest values (besides for these few high temperature data points - but here it might be reflected that some samples collected more air volume than others), so your statement here does not reflect that correctly. I am not even sure if the discrimination between MA and AD makes sense? Couldn't there also be dust particles for some of the filters collected during MA? The one trajectory shown in the SI doesn't say much about this.

line 511: Why is the trajectory shown for BB in the SI so short? If African dust would have been sampled during BB, that could dominate what you see. Alternatively, it could explain that the data is quite low if BB would not have contributed much to INP and the air mass did not come from Africa. A longer trajectory can help the discussion.

line 519: "(likely southern Mexico and Central America)" again made me wonder how you defined periods which were included in BB.

equation 3 and related text: Was the total surface area based on the aerodynamic diameter at the sampling RH used, here? From the parameter-names you use in the main text and in the SI, it seems that this is not the case? Please explain all this in more detail, at least in the SI. If the aerodynamic diameter at the sampling RH was used to calculate n_s, then why? Typically, dry particle sizes are used. It is not described anywhere, why all these transformations are given in the SI for the calculation of surface active site density (n_s) (aerodynamic to geometric, dry to RH). This certainly has to do with your optical size measurement, but all of this needs to be made clearer.

line 541ff, related to Fig. S4: Umo and Grawe only looked at ash particles, so their n_s relates to these, only. You include the surface area of all other particles, too, therefore it is logical that you end up with lower values. This should be said somewhere explicitly

in order not to confuse the readers. Something similar is true for most of the studies you used here for comparison: They examined a separate substance. Even Price only used the surface area of the (massive) amounts of dust particles, if I am not mistaken. It therefore makes perfect sense that mainly Gong fits as all others did not determine the n_s of an air mass with a mix of particles in it, but the n_s of one particle type. Also: are these separate components (starting with montmorillonite) your own measurements? Or else where do they come from? Adjust the text in the main manuscript accordingly!

line 582-583: As said above, INP concentrations of MA and AD were very similar as presented in Fig. 7 (as said, do not overinterpret the onset-temperatures), while indeed n_s was different. But are the INP in MA and SD clearly different? It's fine if this can be shown, but after going through the reviews, please adjust the text here accordingly.

Technical comments:

line 39-40: Add the temperatures at which the INP concentrations given here are valid - without that, it cannot be judged if these are high or low.

line 61: Change "its" to "their".

line 78: "Augustin-Bauditz et al. (2008)": I checked, as the date struck me as quite early, but this is a publication from 2014. This study cited here also focusses on K-feldspar as being the main mineral that drives ice nucleation, so it could additionally fit to what is said in line 84. (Discovery of this slip caused that I randomly checked other citations as well - Welti et al. (2020) should be cited as given below, and in the SI, at least Grawe et al. (2016) is missing in the References. - There may be other issues, please check.)

line 91-92: The samples for this study were not taken on Cape Verde, but flying over the ocean around Cape Verde, with some flights closer to the African continent. Maybe say "... and -23°C for airborne samples collected under dusty conditions around Cape Verde, Africa , ...".

line 94-95: If you follow the above advise, then here “in the same region” has to become "on one of the Cape Verde islands."

line 120: Delete the second “the” (at “in the Arctic the”).

line 163: Add “of the role” between “understanding” and “that”.

line 190: Concerning “20°C and 34°C”: What are these two different temperatures (different seasons? day and night?)? Why are there two values here while above it was only one value for Merida and the Yucatan peninsula?

line 191: For the RH, the "+" part here makes no sense, as that ends up well above 100%.

line 196: Exchange “for presented” with either "for presentation" or "to be presented".

line 228: Better use “consists of” instead of “is integrated by”.

line 230 and line 232 and line 236: cmˆ3 (not only cm)!

line 235: Where is this thermocouple? Maybe explicitly hint to Fig. S1 here, too.

line 251: How is the air “generated”? Maybe better: "added to the nitrogen flow when it is conducted ...".

line 255: Do you know the temperature variation across the glass plate? Please add this information here.

line 261: Be careful with the time form you use - this whole paragraph is in present tense, besides for this "were", here. -> are.

line 261: Instead of “with” better “on” or “in”.

line 296: Either " diameters ranging from ... to ..." or "diameters between . . . and . . .".

line 297: Give the size range for which these two LasAir-instruments measured, in the text or table.

line 325: Why do you give two different lengths for the trajectories? Different for different samples / locations/ . . . ? Explain this in the text.

line 402: Replace “areas” with “parts”.

line 402: Add “is” prior to “positively”.

line 404: Add "a comparatively high mass fraction of" between “contained” and “particles”.

line 427: Fig. 6 shows the concentration, not the distribution -> correct.

line 487: At least mention briefly, what the explanation is, that is given for your observation in the here cited publication.

line 558: Concerning “mixed-phase cloud formation“: this should better be “immersion freezing” - you do not mimic the formation of a cloud, that would be up to cloud chambers.

line 561-564: As that PICNIC-study is not a topic described in your manuscript (besides that you mentioned that this other study exists), this should not be part of these conclusions here. Sentence should be deleted.

line 576: Concerning “and also at tropical latitudes.” - How about Gong et al. (2020), which you’ve cited a number of times?

Fig. 4: In the caption: “Khon” -> “Kohn”.

Fig. 7b: Black writing in a dark blue field is quasi invisible - change color of the text to white.

SI, Fig. S3: Typically the sequence you used so far is "MA", "BB", "AD" (following the sequence with which you sampled), so it is a bit confusing that the latter two are swapped, here. Please exchange.

SI, line 89: “base” -> “based”.

Literature:

Augustin-Bauditz, S., H. Wex, S. Kanter, M. Ebert, F. Stolz, A. Prager, D. Niedermeier, and F. Stratmann (2014), The immersion mode ice nucleation behavior of mineral dusts: A comparison of different pure and surface modified dusts Geophys. Res. Lett., 41(20), 7375-7382, doi:10.1002/2014GL061317.

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Wex, H., L. Huang, W. Zhang, H. Hung, R. Traversi, S. Becagli, R. J. Sheesley, C. E. Moffett, T. E. Barrett, R. Bossi, H. Skov, A. Hünerbein, J. Lubitz, M. Löffler, O. Linke, M. Hartmann, P. Herenz, and F. Stratmann (2019), Annual variability of ice nucleating particle concentrations at different Arctic locations, Atmos. Chem. Phys., 19, 5293–5311, doi:10.5194/acp-19-5293-2019.

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