

We would like to thank the Editor and the Reviewers for their additional constructive suggestions, which helped us to improve the manuscript. Specific answers and manuscript modifications related to the Editor and Reviewers comments are given below in red text.

### **Editor**

(1) Referee 2 comment 1: I agree with referee 2 that it appears somewhat unusual to define the three types of aerosols (marine aerosol, MA, biomass burning, BB, and African dust, AD) by the time period of sampling, rather than by their chemical composition or by their origin (via back-trajectory analysis). I do understand that there is a correlation between those classifications, but from the data provided in the current manuscript, it is not clear to me how strong this correlation is. Such information is required! Moreover, you have defined background aerosol number and composition. Apparently, these are distinctly different from the general classification aerosol properties (see Figs5b and 5c in particular). So my question: Is the background day just one unusual day within the time-period otherwise influenced by the general aerosol according to its classification, or do “background days” occur more often? Note that because of the high non-linearity of ice-nucleation activity between different types of aerosol, the time-period averaging will strongly depend upon the fraction of days with aerosols of the general classification during that period. It will also depend upon the fraction of INP measurement days dominated by that particular class. For that purpose, I suggest to make that classification based on chemical analysis for the days of INP measurements or, if this is not possible because of non-sufficient overlap between days with chemical analysis and days of INP measurement, based on back trajectories for the INP measurement days. Then do provide the numbers for the fractions of days in each period at least. Moreover, I appreciate the addition of supplementary table S1. However, even when considering both Table 1 and Table S1, I found myself struggling on which days INP data, size distribution data, and chemical analysis data were obtained. A more detailed overview table with individual days of INP measurements, days of chemical analysis measurements and the definition of aerosol classifications would help. All readers interested in further detail can then have a look at the original data of the individual analyses and INP measurement in the deposited original data, see point (7) below.

A/ We thank the Editor and the Reviewer for pointing out the lack of clarity in the discrimination between the three air masses and the meaning of the backgrounds.

Prior knowledge of the seasonality of BB and AD events was crucial to select the periods to carry out the field campaigns. As shown below in Figure A1, April is the time of the year where the maximum fire density is found in the Yucatan Peninsula. Note, that these results were obtained from a 14-year study using satellite information (Rios and Raga, 2018).

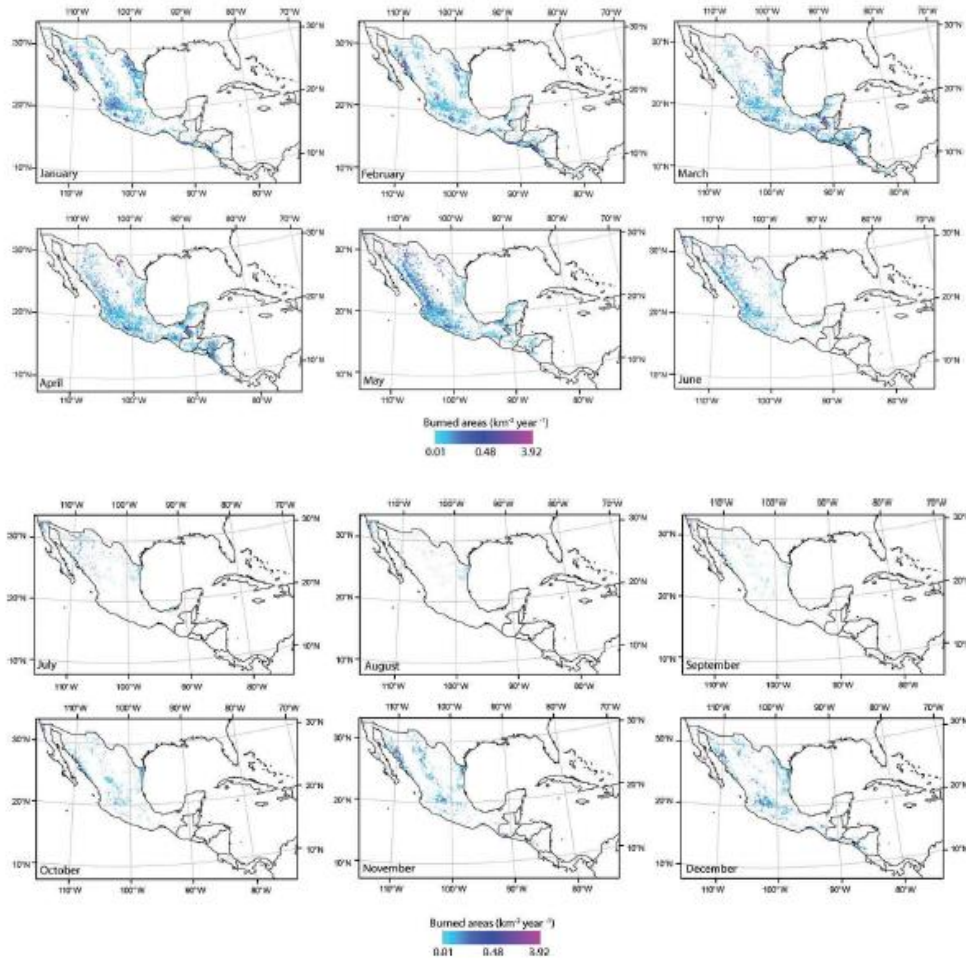


Figure A1. (Top) Spatial distribution of the 14 year average burned area ( $\text{km}^2$ ) in Mexico and Central America for the period (2001–2014) between January and June. (Bottom) Spatial distribution of the 14 year average burned area ( $\text{km}^2$ ) in Mexico and Central America for the period (2001–2014) between July and December (Ríos and Raga, 2018).

Likewise, Figure A2 shows that July has the highest probability of dust arrival in the Yucatan. Moreover, July is characterized in many regions around the Caribbean as part of the mid-summer drought, when there is a reduction in precipitation and stronger trade winds and the presence of the Caribbean Low Level Jet, which has been studied since the 90s (e.g. Magaña et al, 1999). Note that Figure A2 was obtained from 20 years of reanalysis using MERRA-2.

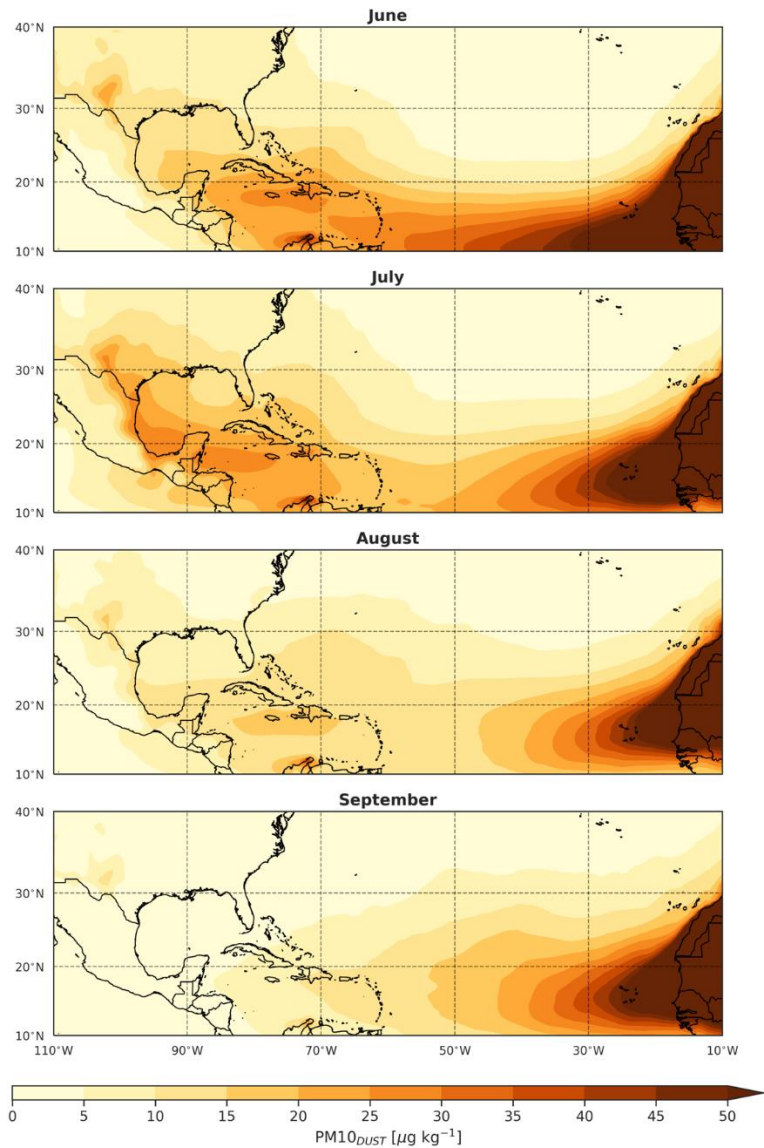


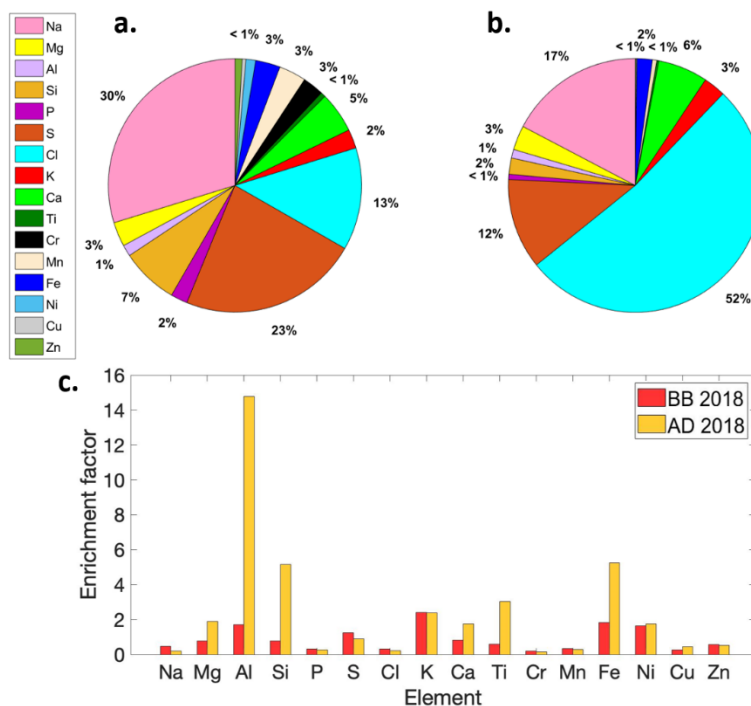
Figure A2. Mean monthly PM<sub>10</sub> concentration associated with dust derived from MERRA-2 over 2000-2019, for June through September. The Yucatan peninsula is located in southeastern Mexico, surrounded by the western Caribbean to the East and the Gulf of Mexico to the North and West (Raga et al. 2020).

To clarify how the MA, BB, and AD periods were chosen, the following text was added to the revised manuscript. Lines 216-228: “*The presence of BB particles and the intrusions of AD onto the Yucatan Peninsula has been previously documented (e.g., Yokelson et al. 2009; Kishcha et al. 2014; Rios and Raga, 2018; Raga et al. 2020; Trujano-Jiménez et al. 2021; Ramirez-Romero et al. 2021). Rios and Raga (2018) reported that within the BB season, the maximum fire density is observed in April. Likewise, Kishcha et al. (2014), Raga et al. (2020), and Ramirez-Romero et al. (2021) indicate that July is the period with the highest likelihood for the AD influx to the Western Caribbean within the (MSD). Therefore, April and July were chosen as the sampling periods to capture the presence of BB and AD particles in Mérida, respectively.*

*Given that the Yucatan Peninsula is encircled by the GoM, MA is ubiquitous throughout the Peninsula. Therefore, the MA composition was assessed in the remote coastal village of Sisal between January and February, a time of the year where the presence of particles such as BB and AD are least likely.”*

Having in mind that the aerosol particles in the Yucatan Peninsula in January, April, and July are heavily influenced by MA, BB, and AD, respectively (based on the literature), we analyzed the aerosol’s chemical composition and the history of the air masses, to corroborate their origin.

Regarding Figure 5, we would like to mention that the original figure showed the elemental composition for several days, from each period, as examples for typical days. The background compositions were added to left of those panels in order to highlight the differences for the readers. However, as the figure has created confusion, we have modified it (as shown below) and subsection 3.2 was rewritten (Lines 431-498). The redrawn Figure 5 shows the background composition by site rather than by sampling period to avoid confusion and the influence of the BB and AD in the aerosol composition was assessed by adding the enrichment factor for each of the 16 analyzed elements.



**Figure 5.** Percentage elemental mass concentration for background samples in Merida (panel a) and Sisal (panel b). Enrichment factors are calculated for samples obtained under the influence of BB (red bars) and AD (yellow bars) plumes from background conditions in Merida (panel c).

As mentioned above, the data provided in the original Figure 5 corresponded to some example days and not to the full data available. We may have given the impression that we only have the chemical composition for limited days; however, we would like to clarify that the chemical composition was obtained for >90% of those days where the INP concentrations are being reported in the present study. We hope that the new Figure 5 (and the corresponding text) has alleviated the reviewer's and editor's concerns.

The aim of Figures 5 and S3 was to provide robust evidence that the aerosol particles collected in January, April, and July were different in origin. In parallel to the aerosol particles collected to analyze their chemical composition, aerosol particles were collected with the MOUDI to evaluate their ice nucleating abilities. Therefore, we have INP measurements for each sampling period together with chemical composition and size distribution as shown below in the revised Table S1.

**Table S1.** Summary of a subset of samples (those with the full size range available i.e., from 0.32  $\mu\text{m}$  to 10  $\mu\text{m}$ ) taken from Merida and Sisal during 2017 and 2018 to analyze the results presented in this study. MA, BB, and AD refer to marine aerosol, biomass burning, and African dust, respectively. The last two columns indicate if the chemical composition and the size distributions were available parallel to the INP samples. \*two samples were collected at different times during the same day.

Aerosol Type	Place	Date	Chemical composition available	Size distribution available
MA	Sisal	24-01-2017 *	Yes	Yes
		24-01-2017 *	Yes	Yes
		25-01-2017	Yes	Yes
		26-01-2017	Yes	Yes
		27-01-2017	Yes	Yes
		28-01-2017	Yes	Yes
		29-01-2017	Yes	Yes
BB	Merida	27-05-2017	No	No
		03-04-2018	Yes	Yes
		05-04-2018	Yes	Yes
		07-04-2018	Yes	Yes
		08-04-2018	Yes	Yes
AD	Sisal	11-07-2018	Yes	Yes
		12-07-2018	Yes	Yes
		13-07-2018	Yes	Yes
		15-07-2018	Yes	Yes
	Merida	11-07-2018	Yes	No
		13-07-2018	Yes	No
		14-07-2018	Yes	Yes
		16-07-2018	No	No

(2) Referee 2, comment 2: “Only one in  $10^5$  to  $10^6$  of the aerosol particles can act as INP at temperatures higher than  $-38^\circ\text{C}$  (Lohmann et al., 2016).” I agree with you that this is a cited statement from the given reference. Considering the comment by the referee, I suggest that you may consider making this sentence less strict and approximate.

*A/ Lines 69-70 were changed as follows “One in  $10^6$  (or fewer) of atmospheric aerosol particles can act as INP at temperatures higher than  $-38^\circ\text{C}$ ”.*

(3) Referee 2, comment 3: I agree with the suggestion of the referee for changing the text to: “This is the first such comprehensive study ever conducted in Mexico and among the first ones at tropical latitudes.” Or something similar.

*A/ Thanks for your suggestion. The text was modified as follows. Lines 659-660: “This is the first such comprehensive study ever conducted in Mexico and among the first ones at tropical latitudes.”*

(4) Referee 2, comment 4: I agree with the referee that a statement on storage time of the samples should be added to the experimental part, and in that one you may also make a statement, as you did in your previous answer to the referee, that you did not observe differences between 12 and 24 months of storage. Moreover, the current study was performed before the Beall et al. (2020) study on protocols for the storage of INP samples became publically available, and it was submitted before the latter was accepted for final publication. Therefore, I think it suffices to only briefly mention that study in that respect.

*Thank you for your suggestion. The text was modified as follows. Lines 245-250: “After each sampling, the glass coverslips were stored in Petri dishes between three and twenty-two months at  $4^\circ\text{C}$  prior to their analysis with the DFT in Mexico City (Fig. 2a). As recently highlighted by Beall et al. (2020), the temperature and the length of the storage can impact the ice nucleation abilities of MA samples. Although, this was not evaluated in the present study, future studies will evaluate how the storage procedure impacts results.”*

(5) Referee 2, comment 5: While the referee is correct in their statement, I think it is okay if you leave the data by Umo et al. (and other data sets) in the comparison figures. But I would ask you to add a statement on that account to inform readers, i.e. “Note that some of the  $n_s$  data in the comparison refer to pure components or aerosol types, while our analysis includes the entire atmospheric aerosol variability, i.e. also the surface on particles not acting as ice nucleating particles” or something along this line.

*A/ Thank you for the suggestions. The following text was added to the revised manuscript. Lines 613-615 “Note that some of the  $n_s$  data in the comparison refer to pure components or aerosol types, while our analysis includes the entire atmospheric aerosol variability, i.e., also the surface of particles not acting as ice nucleating particles.”*

(6) Referee 2, comment 6: I agree with the referee, so please add the information on how the size distribution for calculating the  $n_s$  values were obtained/calculated so that it can be clearly understood and repeated.

*A/ This section was modified to include the requested information as follows:*

### **“Calculation of surface active site density ( $n_s$ )**

The methodology employed in this study is based on Si et al. (2018). First, the particle's density was calculated at a given RH ( $\rho_{p,RH}$ ) using Equation S\_E1. Later on,  $x$  factor was calculated following the Equation S\_E2. Note that the  $x$  factor was computed for each air mass type (MA, AD, and BB). Finally, to obtain the  $n_s$  values using Equation S\_E3, the particle concentration as a function of their size measured with LasAir and the INP concentrations were necessary. The  $n_s$  was obtained for each individual sample listed in Table S1, and therefore, the INP concentration and the particle concentration of each individual sample (and for each period) were used.

#### **1. Calculation of the particle density at a given RH ( $\rho_{p,RH}$ )**

$$\rho_{p,RH} = \rho_w + (\rho_{p,dry} - \rho_w) \frac{1}{gf^3} \quad (\text{S\_E1})$$

where  $\rho_w$  is the density of water and  $\rho_{p,dry}$  is the density of the dry particles.  $1.87 \text{ g cm}^{-3}$  was used for marine aerosol (Si et al., 2018),  $2.5 \text{ g cm}^{-3}$  for mineral dust particles (Wheeler et al., 2015) and  $1.25 \text{ g cm}^{-3}$  for biomass burning particles, as it is the average between  $1.1 \text{ g cm}^{-3}$  and  $1.4 \text{ g cm}^{-3}$  reported by Li et al. (2016).  $gf^3$  is the hygroscopic growth factor that was obtained from Ming and Russell (2001), using the mean relative humidity for Sisal in January (65%) and July (95%), and for Merida in April (65%) and July (90%). The particles were assumed to be composed of 30% of organic species.

#### **2. Calculation of factor ( $x$ ).**

$$x = gf \sqrt{\frac{\rho_{p,RH}}{\chi \rho_0}} \quad (\text{S\_E2})$$

where  $\chi$  is the dynamic shape factor for a non-spherical particles and  $\rho_0$  the unit density of  $1 \text{ g cm}^{-3}$ .

#### **3. Calculation of $n_s$ based on the geometric diameters at a given RH.**

$$n_{s,ae,RH} = \frac{[INP]}{S_{tot,ae,RH}} = \frac{[INP]}{\pi x^2 D_{geo,dry}^2 N_{tot}} \quad (\text{S\_E3})$$

where  $[INPs]$  is the concentration of INP ( $L^{-1}$ ) at each temperature (i.e.,  $-15^\circ\text{C}$ ,  $-20^\circ\text{C}$ ,  $-25^\circ\text{C}$ , and  $-30^\circ\text{C}$ ) for each sample.  $S_{tot,ae,RH}$  is the total surface area based on the aerodynamic diameter at the sampling RH, and  $N_{tot}$  the total number of aerosol particles.  $D_{geo,dry}$  corresponds to the average diameter of each LasAir size bin as shown in Table S2. In this case, only the data from channels 1 to 4 from the LasAir were used since these size range overlaps with the MOUDI diameters from stage 2 to 7.

**Table S2.** LasAir channels and diameters used in the present study with their corresponding surface area.

<i>LasAir channel</i>	<i>Diameter range (<math>\mu\text{m}</math>)</i>	<i>MOUDI stages (<math>\mu\text{m}</math>)</i>	<i><math>D_{\text{geo, dry}} (\mu\text{m})</math></i>	<i><math>D^2_{\text{geo, dry}} (\text{cm}^2)</math></i>
<i>D1</i>	<i>0.3 – 0.5</i>	<i>7 (0.32-0.56)</i>	<i>0.4</i>	<i><math>1.60 \times 10^{-9}</math></i>
<i>D2</i>	<i>0.5 – 1.0</i>	<i>6 (0.56-1.00)</i>	<i>0.75</i>	<i><math>5.62 \times 10^{-9}</math></i>
<i>D3</i>	<i>1.0 – 5.0</i>	<i>3+4+5 (1.00-5.6)</i>	<i>3.0</i>	<i><math>8.99 \times 10^{-8}</math></i>
<i>D4</i>	<i>5.0 – 10</i>	<i>2 (5.6-10.0)</i>	<i>7.5</i>	<i><math>5.62 \times 10^{-7}</math></i>

For each sample listed in Table S1, the INP concentration was derived. Also, given that the LasAir continuously measures the particle size distribution (PSD), the total particle concentration ( $N_{\text{tot}}$ ) for each LasAir size bin listed in Table S2 was derived for exactly the same time interval of each INP sample.

As shown in Table S2, the INP concentration from the MOUDI stages 3, 4, and 5 were combined in order that their sizes match those from the LasAir size bins when calculating the  $n_s$  values for each sample.”

(7) As this manuscript is being considered as a measurement report, the original data of the study has to be made available, e.g. either as data sets in the supplement, or deposited and archived in public databases (with doi or hyperlink given): “The data presented in measurement reports must be openly accessible in accordance with the EGU data policy.” See: [https://www.atmospheric-chemistry-and-physics.net/about/manuscript\\_types.html](https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html) The data policy statement includes: “Therefore, Copernicus Publications requests depositing data that correspond to journal articles in reliable (public) data repositories, assigning digital object identifiers, and properly citing data sets as individual contributions. [...] In addition, data sets, model code, video supplements, video abstracts, International Geo Sample Numbers, and other digital assets should be linked to the article through DOIs in the assets tab.” and can be found here: [https://www.atmospheric-chemistry-and-physics.net/policies/data\\_policy.html](https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html) A very recent example that was published yesterday is shown here: <https://acp.copernicus.org/articles/20/15969/2020/>

A/ As discussed in the Ice Nucleation Colloquium presented in January 12, 2021, there is a paucity of ice nucleation data from Tropical latitudes and we have been working hard to partially fill this lack of data and to build a unique data set in this direction. Our next step is to develop a “Tropical” ice nucleation parametrization to be implemented in a regional climate model where it can be compared against the currently available ice nucleation parametrizations which mostly contain mid- and high-latitude measurements. Therefore, we are not comfortable making our unique data set available to the wider community before we have developed the aforementioned parametrization. While we are open to sharing our data with interested researchers, we kindly request the Editor to allow us to share our data upon request to the corresponding author in order that we are informed on how our measurements will be used.

(8) Please consider the technical/minor points raised by referee 2.

A/ The technical points were corrected.



(9) Please correct all the technical points raised by referee 1.

A/ The technical points were corrected.

### **Reviewer #1**

**L121:** “Mccluskey” should be “McCluskey”. Please change along the text.

A/ Thank you. This was corrected along the text.

**L138:** Remove one space after the word: “Vancouver”

A/ Corrected.

**L246:** A space is missing between cm and x.

A/ Corrected.

**L537:** “eastern” should be “Eastern”

A/ Corrected.

**L640:** LL is LAL?

A/ Corrected.

### **Reviewer #2**

The manuscript improved some, and as much as I would like to give the article a “go”, I still cannot do this.

The following issues cannot be left uncommented as they are wrong or inconsistent. In the end, the Editor will have to decide, to which extent the authors will have to review their manuscript once more. In any case, below are my remaining concerns, which were mentioned before and not regarded in the revised version.

Therefore I still have to say “major revisions”. Most issues can probably be revised with small additions, but with my choice of “major revisions” I want to stress that these changes are really needed and should not be swept aside.

In the following, line numbers refer to those of the following file: “acp-2020-783-author\_response-version1.pdf

**1** One of the shortcomings I still see is the way how the data were split into three different periods, referred to as “marine aerosol” (MA), “biomass burning” (BB) and “African dust” (AD). This needs to be explained and motivated somewhat better in the text. I mentioned that before, but as it was not done, I elaborate here, why that should be done, and how the current text is not sufficient:

For each period, it is assumed that INP for each day/sample are representative for the whole period, as no further discrimination is done. In the answers to my previous review, you repeatedly refer to Figs. 5 and S3. These figures show elemental composition for 3 different days for each period (one always referred to as “background”) and 3 single back-trajectories, one for each period, respectively.

A/ We thank the reviewer for pointing out the lack of clarity in the discrimination between the three air masses and the meaning of the backgrounds.

Prior knowledge of the seasonality of BB and AD events was crucial to select the periods to carry out the field campaigns. As shown above in Figure A1, April is the time of the year where the maximum fire density is found in the Yucatan Peninsula. Note, that these results were obtained from a 14-year study using satellite information (Rios and Raga, 2018).

Likewise, Figure A2 (above) shows that July has the highest probability of dust arrival in the Yucatan. Moreover, July is characterized in many regions around the Caribbean as part of the mid-summer drought, when there is a reduction in precipitation and stronger trade winds and the presence of the Caribbean Low Level Jet, which has been studied since the 90s (e.g. Magaña et al, 1999). Note that Figure A2 was obtained from 20 years of reanalysis using MERRA-2.

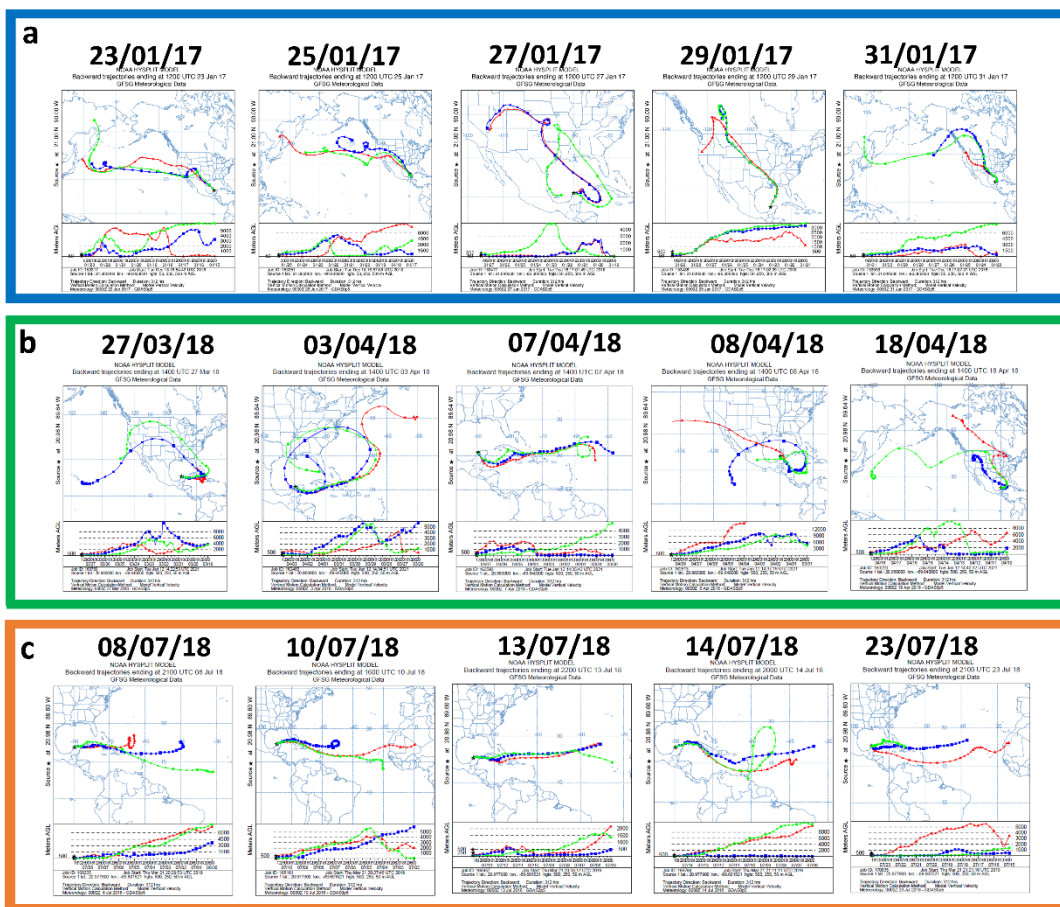
To clarify how the MA, BB, and AD periods were chosen, the following text was added to the revised manuscript. Lines 216-228: *“The presence of BB particles and the intrusions of AD onto the Yucatan Peninsula has been previously documented (e.g., Yokelson et al. 2009; Kishcha et al. 2014; Rios and Raga, 2018; Raga et al. 2020; Trujano-Jiménez et al. 2021; Ramirez-Romero et al. 2021). Rios and Raga (2018) reported that within the BB season, the maximum fire density is observed in April. Likewise, Kishcha et al. (2014), Raga et al. (2020), and Ramirez-Romero et al. (2021) indicate that July is the period with the highest likelihood for the AD influx to the Western Caribbean within the (MSD). Therefore, April and July were chosen as the sampling periods to capture the presence of BB and AD particles in Mérida, respectively.*

*Given that the Yucatan Peninsula is encircled by the GoM, MA is ubiquitous throughout the Peninsula. Therefore, the MA composition was assessed in the remote coastal village of Sisal between January and February, a time of the year where the presence of particles such as BB and AD are least likely.”*

Having in mind that the aerosol particles in the Yucatan Peninsula in January, April, and July are heavily influenced by MA, BB, and AD, respectively (based on the literature), we analyzed the aerosol’s chemical composition and the history of the air masses, to corroborate their origin.

Regarding Figure 5, we would like to mention that the original figure showed the elemental composition for several days, from each period, as examples for typical days. The background compositions were added to left of those panels in order to highlight the differences for the readers. However, as the figure has created confusion, we have modified it (as shown below) and subsection 3.2 was rewritten (Lines 431-498).

Also, Figure S3 was modified to include additional days. The initial conclusions did not change, as the trajectories for the three air masses come from different locations. We would like to make clear that the sampling period and the assigned labels MA, BB, and AD are not based on the back-trajectories alone. As explained above, this is based on literature data and previous observations. Therefore, the HYSPLIT back-trajectories were not the sole indicator of air mass source, but rather were complementing a variety of aspects considered jointly.



**Figure S3.** 13-day HYSPLIT back trajectories for the three different air masses for (a) MA-2017, (b) BB-2017, and (c) AD-2018.

However, days for which you show the chemical composition are often not those days for which INP samples were included. For example, of the 7 INP samples you use for MA, the chemical composition is given for two days which were also used for INP analysis, but not for the third. And of the two that are shown one is the background day. For BB and AD, of the three days for which chemical compositions are shown, for each period only one appears among the INP samples.

A/ As mentioned in our previous answer, the data provided in the original Figure 5 corresponded to some example days and not to the full data available. We may have given the impression that we only have the chemical composition for very few days; however, we would like to clarify that the chemical composition was obtained for >90% of those days where the INP concentrations are being reported in the present study (revised Table S1). We hope that the new Figure 5 (and the corresponding text) and the revised Table S1 has alleviated the reviewer concerns and clarified how the decision was made to select specific time periods for sampling.

Also, for these single trajectories that are only shown for each phase (MA, BB and AD), the one single trajectory for BB and that single one for AD are given for days for which no sample is included in the INP analysis. And the trajectory you show for MA is that for the background conditions.

A/ Similar to the original Figure 5, the original Figure S3 showed examples of typical back-trajectories for each sampling period. We have modified Figure S3 to add other days including those where the INPs analysis is being reported.

And what does the background condition mean, anyway? This is nowhere clearly defined (with that I aim at what “background” means with respect to the aerosol during these phases, not that these are the days with the lowest mass concentrations.)

A/ New Figure 5 shows the background composition by site and not by sampling period to avoid confusion. Section 3.2 was rewritten and it includes an explanation on how the backgrounds are defined (Lines 432-499).

It is, therefore, still unclear to me how representative the INP samples you chose are for the conditions during each of the three different phases. This cannot be judged based on these few data you show in Figs. 5 and S3.

A/ The aim of Figures 5 and S3 was to provide robust evidence that the aerosol particles collected in January, April, and July were different in origin. In parallel to the aerosol particles collected to analyze their chemical composition, aerosol particles were collected with the MOUDI to evaluate their ice nucleating abilities. Therefore, we have INP measurements for each sampling period together with chemical composition and size distribution as shown below in the revised Table S1. Although our data set is larger than the listed samples in Table S1, in order that the provided analysis was comparable for the different field campaigns, we did not take into account those samples where the information from “all” MOUDI stages was not available (i.e., from stage 2 (10  $\mu\text{m}$ ) to stage 7 (0.32  $\mu\text{m}$ )).

So overall, there is this inconsistency that you treat all days during one period similar when it comes to interpreting INP, but on the other hand you give “background conditions” that are clearly different from other days in the same season. And nowhere is there a complete overview over the conditions for all days for which INP samples were used.

A/ We hope that with the new Figure 5 and the rewritten subsection 3.2 these points were clarified.

Such an overview could quite simply be done if backward trajectories would be given for all days for which INP samples are included, and that is what I recommend you to do.

A/ As mentioned above, Figure S3 now includes additional back-trajectories for each sampling period. However, we would like to stress that the HYSPLIT back-trajectories were not the sole indicator of air mass source, but rather were complementing a variety of aspects considered jointly.

It is also important to mention that our setup (i.e., the UNAM-MOUDI-DFT) is not a high temporal resolution INP sensor as we need to run the MOUDI for >4 h to collect one sample. Therefore, during the >4 h sampling period the origin of the air masses arriving to the sampling site might change. Based on the literature data (see above Figures A1 and A2), the chemical composition of the MA, BB, and AD shown in new Figure 5, and their size distributions (Fig. 6) we are confident that we were sampling aerosol particles from completely different origins in January, April, and July. Thus, we evaluated if those differences in the origin of the aerosol particles, i.e., MA, BB, and AD impacted their ice nucleation abilities as shown in Figures 7, 8 and S4.

Along the same line, it makes no sense to say that this one 24 hour backward trajectory suffices for the BB phase (which is what you answer to my review, but also still say in line 1386 “The 24 h back trajectories were run for the BB events as they were likely locally emitted.”); because you later claim, concerning the BB phase, in lines 1589-1591 “The lower concentrations reported in the present study can be attributed to the long distance between the burning areas (likely southern Mexico and Central America) and the sampling site.” If that latter is correct (which it may very well be, you cannot exclude this), then the 24 h back-trajectories you showed are too short. If you do the backward-trajectories for all INP samples, use the longer time period for all!

Then, depending on how these trajectories are, the text may have to be adjusted. And if not much adjustment is needed, all the better. But then at least your data put into a much stronger context.

*A/ Please see our previous answer.*

**2** Line 1073: What really irritated me is, that you write that you understood my concern about the statement “Only one in  $10^5$  to  $10^6$  of the aerosol particles can act as INP at temperatures higher than  $-38^\circ\text{C}$  (Lohmann et al., 2016).”, but that you did not want to change it. When something is wrong in literature, it should not be promoted further (even if others have done that before). Agreeing that this statement is wrong but insisting to use it anyway is rather strange to me, particularly as it does not influence anything in your work. Why not saying it correctly? DeMott et al. (2016) more correctly say “... INPs ..., a select subgroup that may represent 1 in  $10^6$  or fewer of all aerosol particles”. Not perfect, but better.

*A/ Lines 69-70 were changed as follows “One in  $10^6$  (or fewer) of atmospheric aerosol particles can act as INP at temperatures higher than  $-38^\circ\text{C}$ ”.*

**3** Along a similar line as my last comment goes the following, now in lines 1649-1650, “This is the first such comprehensive study ever conducted in Mexico and also at tropical latitudes.” I commented that this is not true for tropical latitudes as Gong et al. (2020), a study you cited several times and thus should know about, also measured at tropical latitudes. Your answer was “Gong et al. (2020) was indeed performed at Tropical Latitudes; however, the authors focused on a single aerosol type (MA), instead of three different and distinct air masses as in the present study.”

So now we would need to define the word “comprehensive”, but in my understanding you are saying “Yes, our statement is not true, but we will make it anyway as this other study did not study the exact same thing as we did.”

Additionally, as you say, by now also Welti et al. (2020) is published and includes data for tropical latitudes. I would suggest you change the wording of the respective sentence at least to: “This is the first such comprehensive study ever conducted in Mexico and among the first ones at tropical latitudes.”

*A/ The text was modified as follows. Lines 659-660: “This is the first such comprehensive study ever conducted in Mexico and among the first ones at tropical latitudes.”*

**4** Line 1255: Different from what you say, it has been discussed in the community already for quite some time that samples need to be stored frozen. It has even been shown that long storage times under freezing conditions may change a bacterial sample (Polen et al., 2016). It now was reported that storage at  $-20^\circ\text{C}$  is mandatory (Beall et al., 2020). In your answer to

my first review, you compare two groups of samples which were both stored at 4°C for many months (12 and 24), and their similarity might just mean that both degraded to the same degree, and that after many months of storage at this comparably high temperature of 4°C no further changes occur. At least cite and shortly comment on the new study, as this very well could influence the outcome of your study. And consider changing your procedures in the future.

A/ Thank you for your suggestion. The text was modified as follows. Lines 245-250: *“After each sampling, the glass coverslips were stored in Petri dishes between three and twenty-two months at 4°C prior to their analysis with the DFT in Mexico City (Fig. 2a). As recently highlighted by Beall et al. (2020), the temperature and the length of the storage can impact the ice nucleation abilities of MA samples. Although, this was not evaluated in the present study, future studies will evaluate how the storage procedure impacts results.”*

5 line 1616: Again: Umo et al. (2015) looked at only ash particles, so any agreement has to be coincidental, as the  $n_s$  for Umo et al. (2015) relates to the surface area of ash particles, only, while your data relate to total atmospheric aerosol with many other particles and with some ash particles, and also with some soot particles in them! This means in detail: When (at a fixed temperature) Umo et al. (2015) report a value of 1000  $\text{cm}^{-2}$ , then this means that on the surface of the ash particles, there are 1000 INP per  $\text{cm}^2$  that are active at that temperature. When your BB samples show the same  $n_s$ , it means that the BB aerosol has 1000 INP per  $\text{cm}^2$  of the OVERALL aerosol. That is clearly something completely different. Check and correct all of your respective comparisons respectively.

A/ We agree with the reviewer that ambient aerosol particles are complex and that their physiochemical properties may differ from those found in pure substance. Following the Editor’s advice and to acknowledge this, the following text was added to the revised manuscript. Lines 613-615 *“Note that some of the  $n_s$  data in the comparison refer to pure components or aerosol types, while our analysis includes the entire atmospheric aerosol variability, i.e., also the surface of particles not acting as ice nucleating particles.”*

6 Concerning the derivation of  $n_s$ , still more details are needed. If I did not overlook anything, it is nowhere to be found if you used one average surface area from the average size distribution for each of the three phases (MA, BB and AD), and if yes, from which days this average size distribution was constructed (all days for which INP measurements were included?). Or, if the measured size distribution for each separate day was used. At least this information needs to be clearly added. Related is the sentence in line 1498, where you added “(reported by the LasAir)”. First, this inserted new information would be better given in the first sentence of the paragraph. (The sentence you inserted it to is more difficult to understand now). And second, the information you give in the review on how you derived the values shown in Fig. 6 should also be given in the manuscript, so that it becomes clear which value you show in that figure.

A/ This section was modified following the reviewer suggestion, as shown above in the answers to the Editor (point #6).

#### **Minor comments:**

Concerning your method, you answered that direct method comparisons have already been made, citing two papers. Knowing this helps the reader to judge the credibility of your work, and I strongly recommend that you add a short paragraph in which you cite these comparison studies and shortly summarized the respective results for your method.

A/ The following text was added to the revised manuscript. Lines 257-261: *“The results delivered by the Mason et al. (2015) MOUDI-DFT were compared against those reported by the Colorado State University-Continuous Flow Diffusion Chamber (CSU-CFDC). As shown in DeMott et al., (2017), the median INP concentrations from both devices were good agreement.”*

In Lines 421-429 we also briefly described the PICNIC intercomparison campaign where the UNAM-MOUDI-DFT was included. We are not sure if the reviewer is asking us to expand this paragraph further. Given that the detailed information about the PICNIC results will go in a separate manuscript, we do not want to be too detailed here.

**line 1249:** You do not want to describe your method in more details, so as suggested above cite these comparison studies, at least. But still, as readers might want to copy what you do, add at least information on how much does adding this extra plate change in distance in the impactor and as such influence the selected diameters? That may be small but should at least be mentioned, so that others who want to do similar things are aware of the fact that they should use slides as thin as possible.

A/ The following information was added to the revised manuscript. Lines 237-238 *“hydrophobic glass coverslips of 22 mm x 22 mm”*

The MOUDI was traditionally used to collect aerosol particles on different filters to characterize their chemical composition. Each filter (e.g., Teflon, quartz, aluminum, among others) was fixed on each MOUDI stage using a metallic substrate holder provided by the manufacturer. Therefore, we did not add an additional plate to each MOUDI stage. We simply replaced the original substrate holders with new ones that could hold the square glass coverslips.

Finally, as mentioned in our previous answer, additional details about the inter-comparison results were added to the revised manuscript.

**line 1487 ff:** Can it be excluded that these elements come from Saharan dust (during times when this source is weaker or more particles were lost on the way across the Atlantic)? Only then the karstic soil of the region should be taken into consideration. Particularly as you measured close to the ocean, a strong local influence may not be expected.

A/ As explained above, the new Figure 5 shows the enrichment factor of each element. Therefore, we can differentiate between local and external aerosol source.

**line 1555 ff:** You do not want to change these comparisons, which I criticized as being a bit off, and so be it. But at least also mention that the temperatures in air and water are different for the studies done further to the North than for yours, as this may be an important parameter.

A/ Following the reviewer's suggestion, the following text was added to the revised manuscript. Lines 557-559: *“Additionally, given that air temperatures, RH, and sea surface temperatures are considerably different between the Tropics and higher latitudes, these*

*differences could be partially responsible for the latitudinal differences observed in INP concentrations.”*

**line 1577 ff:** As you said above, particles can be lost, and this will likely be the main cause for lower values. This should be repeated here, too, as aging may also contribute, but would show up in a different  $n_s$ , rather than in lower concentrations. Concentrations simply HAVE TO BE lower further away from the source.

*A/ The following text was added to the revised manuscript. Lines 623-629: “Given that AD particles travelled >8000 km before reaching the Yucatan Peninsula, the low  $n_s$  values calculated for the AD particles are not unexpected. It is likely that the most efficient INPs could have been washed out while travelling over the Atlantic. Also, it is well known that aerosol aging can strongly affect the ice nucleating abilities of mineral dust particles (Kanji et al., 2017). Therefore, the composition of the AD particles that arrived at the Yucatan Peninsula may significantly differ from the AD particles found closer to their source, with different ice nucleation efficiencies.”*

**In your answer (line 810) you say:** “We are not comparing the ice nucleating abilities of the three air masses based on the onset freezing values, we did it based on the  $n_s$  values.” But actually, the amount of comparison you do for  $n_s$  values in Section 3.5 is very limited. There are conclusions given on that in the conclusions section which are not given in Section 3.5, which might merit to be included and discussed there.

*A/ The following text was added to the revised manuscript. Lines 631-637: “Large  $n_s$  values can be likely associated with the presence of biological particles as was the case in Si et al. (2018), who linked the high  $n_s$  values with the presence of terrestrial biological particles. However, McCluskey et al. (2018) and DeMott et al. (2016) showed that these kind of particles can also be of marine origin as there is large biological activity in marine environments. Rodriguez-Gomez et al. (2020) showed different terrestrial and marine microorganisms that were identified in the Sisal, those of which can be linked with the high  $n_s$  values found for the MA samples.”*

#### **Technical issues:**

**line 1345:** Delete the “s” in “INP<sub>s</sub>(T)” in equation 3.

*A/ Corrected.*

**line 1458:** Add “in the BB season” after “background conditions”, as this is not the case during MA.

*A/ The text was modified following new Figure 5.*

**line 1461:** Use GoM (instead of Gulf of Mexico), as you’ve done throughout the text.

*A/ Corrected.*

**line 1470ff:** “... corroborating that the sampled air masses during this season contained a comparatively high mass fraction of particles emitted from BB.” At the end of this sentence, it should be added “when PM<sub>2.5</sub> was high”, as you show yourself that potassium was not always high during this phase (Fig. 5 and Fig. S2a).

*A/ The suggested text was added: Line 478.*