

Responses to Reviewer's Comments

We would like to thank the Reviewers for their helpful comments and suggestions, which led to an improved manuscript. Specific answers and revisions to the text and figures related to each reviewer's comments are given below in red bold text.

Reviewer #1

General comments

This manuscript presents data on physical, chemical and biological aerosol parameters observed during a campaign lasting two weeks at a coastal site on the Yucatan peninsula. Technically, the measurements were well done and there are little similar data from the same region. In general, the manuscript is clearly written. The data obtained with the various instruments employed are weakly related, in part because of a mismatch in sampling duration and timing between INP and other parameters (chemical and biological). The sampling duration was per sample 6 hours for INP (2-3 samples a day, morning and afternoon), 48 hours for chemical components 5 min for bacteria and fungi (4 samples a day, in the morning). Further, no link can be made between the cultured microorganisms identified (Tab. 2) and ice nucleation activity, because the cultured organisms were not tested for ice nucleation activity. Much of the relations discussed between INP and other parameters are speculation supported through reference to other literature, without the current study adding substantial new evidence in support of it. Because of that, I would like to encourage the authors to put more effort into relating the different parameters in a way that each parameter can tell us more than its individual story. To start, you could try to combine data from the optical particle counter with size resolved INP concentrations to tell for different size classes the fraction of particles that are ice nucleation active (e.g. what was the ratio INP/aerosol particle in the different MOUDI size classes? How did the ratio change during the passage of the cold front?).

A: We acknowledge the reviewer's careful review and concrete suggestions that have led to an improved manuscript. In particular, the revised version now includes correlations between the chemical composition and the [INP]. Additionally, the [INP] was correlated with the particle number concentration from the LasAir to obtain more robust conclusions (Figure 7 in the revised version).

The influence of cold fronts resulted in a higher fraction of particles acting as INP, especially for particles between 1.0 μm and 5.0 μm in size.

Specific comments

Page 5, line 136-138: Measurements of INP concentrations with the cold cell need to be described in more detail. At least their principle should be clear to reader without having to look up the paper by Mason et al. (2015a).

A: This section has now been expanded to provide additional information of the experimental setup. The following text was added:

Lines 147-154: “The cold cell-microscope system used here is the same one used in previous studies (Mason et al., 2015a,b, 2016; DeMott et al., 2016; Si et al., 2018). The following steps encompass the analysis: i) The samples collected on glass cover slips were placed in the cold cell at room temperature, ii) The cold cell was isolated and kept at 0 °C, while humid air (RH>100 %) was injected into the cell to induce liquid droplets’ formation by water vapor condensation; iii) Dry air (N₂) was then injected into the cold cell to prevent the newly formed droplets from touching. This is a key step to minimize the probability of liquid droplets freezing by contact; and iv) Once droplets’ sizes and thermodynamic conditions were stable, the cold cell was closed.”

Lines 159-174: “The temperature at which each droplet froze was determined by analyzing the video from the CCD camera (XC-ST50, Sony) connected to the microscope and the data reported by the resistance temperature detector (RTD) located at the center of the cold cell with a ±0.2 °C uncertainty (Mason et al., 2015b). Homogeneous freezing experiments were performed on laboratory blanks exposed during the preparation of the MOUDI, while heterogeneous freezing experiments were run on ambient particles deposited on the glass cover slips (Figure S1). The [INP] was calculated using the following expression:

$$[INPs(T)] = -\ln\left(\frac{N_u(T)}{N_o}\right) \cdot \left(\frac{A_{deposit}}{A_{DFT}V}\right) \cdot N_o \cdot f_{ne} \cdot f_{nu,0.25-0.10\text{ mm}} \cdot f_{nu,1\text{ mm}}, \quad (1)$$

where $N_u(T)$ is the number of unfrozen droplets at temperature T , N_o the total number of droplets, $A_{deposit}$ the total area of the aerosol deposit on the hydrophobic glass cover slip, A_{DFT} the area of the hydrophobic glass cover slip analyzed in the DFT experiments, V the total volume of air sampled, f_{ne} a correction factor to account for uncertainty associated with the number of nucleation events in each experiment, $f_{nu,0.25-0.10\text{ mm}}$ and $f_{nu,1\text{ mm}}$ a non-uniformity factor which corrects for aerosol deposit inhomogeneity at a scale of 0.25 - 0.10 mm, and 1 mm, respectively (Mason et al., 2015a). We refer the readers to Mason et al. (2015a) and Mason et al. (2015b) for more details of the MOUDI-DFT operational principle.”

Fig. 3 duplicates the time series measured by the CPC, which is already shown in Fig. 2. Combine Fig. 2 and Fig. 3, and make the time series of wind speed and direction (now panel B in Fig. 3) the top panel of the combined Figure because wind is the factor driving the aerosol concentrations, so logically this factor should come first.

A: Thank you for the suggestion. Figures 2 and 3 have now been combined.

Figure 4: I would like to see the data of the present study as points, not just as a shaded area, where I can not see by how many points a particular part of the area defined. In particular, I am curious to see how many points support the very high [INP] at temperatures above -10 °C. I suggest to revise the Figure in a way that the data taken from Kanji et al (2017) are shown as shaded areas only (no points) and the data of the present study are superimposed on this background as points (perhaps use different symbols for data obtained during the passage of cold fronts).

A: We agree with the reviewer’s suggestion and the figure was completely modified as shown below. This new figure shows the average [INP] as a function of MOUDI stage and also the combined concentration (as grey triangles). The large amount of data from Kanji et al. (2017)

were removed in the revised figure and the revised figure only includes the coastal/oceanic data from DeMott et al. (2016), Welti et al. (2018) and Irish et al. (2019) for comparison.

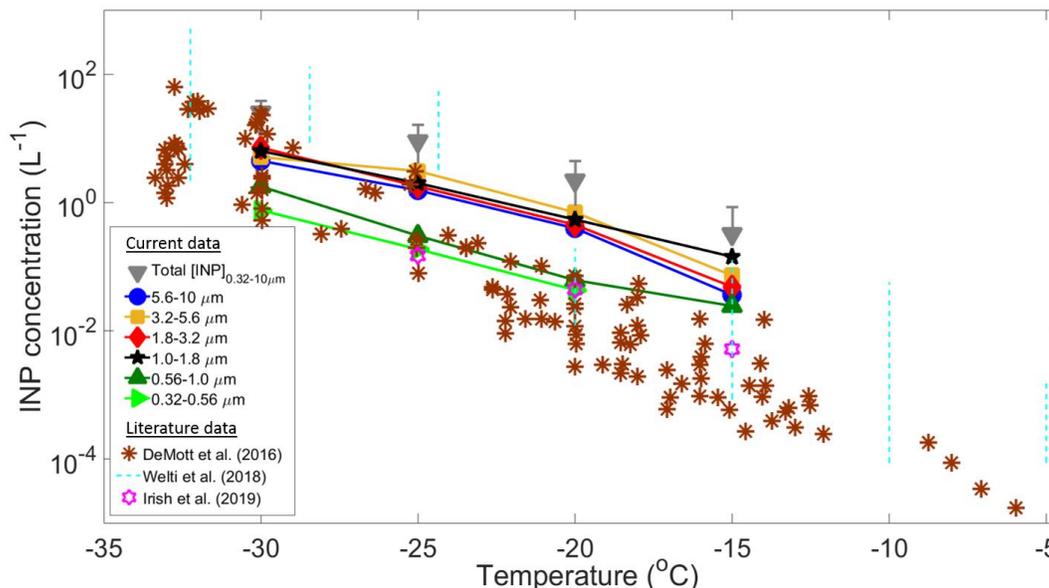


Figure R1. Summary of the INP concentrations as a function of temperature and particle size (solid symbols). Total [INP] are represented by the grey triangles, whereas the brown asterisks, light blue dotted lines, and purple stars are literature data from DeMott et al. (2016), Welti et al. (2018), and Irish et al. (2019), respectively. The upper and lower detection limits of the MOUDI-DFT are 30 L⁻¹ and 0.01 L⁻¹, respectively.

The individual INP scans that corresponded to the shaded area in the old Figure 4 is now presented in the supplementary material (Fig S6). In this improved presentation of the data (Figure 4 and Figure S6) it is clear that particles with sizes between 1.0 μm and 1.8 μm are the most active for temperature above -15°C.

Page 8, first line (and page 11, line 344): The aerosol number concentrations are reported as mean plus-minus one standard deviation, assuming a normal distribution of values. Although this is common practice, it is not correct because aerosol number concentrations have a log-normal distribution. I strongly encourage the authors to apply the less common, but correct metrics (median and multiplicative standard deviation) as explained in Limpert et al. (2001; BioScience, 51, 341-352, freely available at: <https://stat.ethz.ch/~stahel/lognormal/bioscience.pdf>). Why perpetuate a common mistake?

A: We thank the reviewer for this suggestion. The values were adjusted taking into account the log-normal distribution. Lines 262-264: “with geometric mean concentrations (assuming log-normal distributions, Limpert et al. (2001)) for the entire sampling period of 758.51^x/ 1.76 cm⁻³ and 1.00^x/ 1.37 cm⁻³, respectively.”

The Conclusion section is mostly a summary of the Results and Discussion section. It should go further than that.

A: This section has now been modified, given the new analysis performed, and also includes better explanations/conclusions for our results.

Technical corrections

Title: Perhaps replace “importance” with “contribution”

A: The title was changed to “Ice Nucleating Particles in a Coastal Tropical Site”

Page 1, last line: The statement “Biological particles were likely found to be very important. . .” does not make sense to me. Do you mean “Biological particles could potentially be very important.”

A: This sentence was modified.

Page 2, line 37: “Given the potential INP role of a variety of aerosol particles . . .” do you mean “Given the potential role of a variety of aerosol particles as INPs. . .”

Page 3, line 88: “presents”, not “present”

A: Fixed.

Page 3, last line: Why “importance” and not simply “potential relevance”? The word “importance” is a premature value judgement here, at the end of the introduction section.

A: “Importance” was replaced by “potential relevance”.

Page 5, line 128: I would turn the order of the cut-sizes the other way round, so that it follows the logic of the instrument (i.e. 10 um, 5.6 um, . . .0.18 um).

A: As suggested by the reviewer, the order of the cut-sizes was changed.

Page 6, line 162: delete “with”; line 165: replace “0” (zero) in “Na₂CO₃” with “O” (capital “o”).

A: “with” was deleted and Na₂CO₃ was fixed.

Page 8, line 260: change to “At temperatures. . .” (plural).

A: Fixed.

Page 9, line 273: “Saclay”, not “Saclary”.

A: Fixed.

Page 10, line 311: change “elements/cations/ions” to “elements and ions are sodium and chlorine, respectively chloride”; line 322: why “elements/cations/ions” and not just “elements and ions”, cations are ions.

A: Agreed and modified in the revised manuscript.

Page 11, line 353: “temporal mismatch of the data”, not “uncertainty in the analysis”.

A: This paragraph was modified and the sentence was deleted in the revised version.

Reviewer #3

General Comments:

This article discusses INP measurements carried out at Sisal on the coast of the Gulf of Mexico and attempts to identify the sources of INP. They have identified the biological particles from the tropical ocean as the source of INP at measurements site when the wind direction is from the GoM. Considering the importance of the INPs and lack of measurements available over the in tropical latitudes, this article makes a compelling case for the publication. The article is well written although interpretation of measurement can be better. Agreeing with the comment by an anonymous referee, who has commented on the article exhaustively, the authors need to explain the method of INP measurement in brief since that is the main thrust of the article. The lack of correlation between size bin and INP at different temperatures in Fig. 6 warrants detail analysis and in-depth discussion. I would encourage the author to normalize the INP concentration with the total number of particles in respective size bins. Similarly, the chemistry can be analyzed for bins to segregate the contribution of particles of different chemical composition.

A: The correlation between the [INP] and particle size at -20 °C and -25 °C in this study is similar to the results found by Mason et al. (2015b) in the west coast of Canada. However, our results differ at -15 °C and -30 °C and it can partly be attributed to differences in air mass history. This is now acknowledged in the text as follows:

Lines 361-369: "The discrepancies between the present results and those from Mason et al. (2015b) at -15°C and -30°C could be explained by differences in air mass history. Although both studies were conducted at coastal locations, the back-trajectories from the present study indicate that during "normal" days (i.e., 70% of the time) the sampled air masses had a significant continental contribution (Fig. S2). In contrast, air masses were mostly maritime in the Mason et al. (2015b) study. Also, it is important to note that although the cold air crossed the GoM before reaching Sisal, both the US Central Plains and the GoM were likely sources for the aerosol particles present in those cold air masses (Figs. S2B-C and S5)."

The experimental setup to determine the INP concentrations was extended (see above answer to reviewer #1). Additionally, the fraction of aerosol particles acting as INPs as a function of size is now shown in Figure 7.

When correlating the chemical composition and [INP] we found correlation between the average composition per sample with average [INP] per samples as shown in Tables 2 and S1. However, when the correlation is calculated as a function of particle size (per stages), the highest r^2 obtained was just 0.22. It is important to note that these correlations could be misleading given that the XRF reports the mass of the elements (ng m^{-3}) while the MOUDI-DFT reports the number of INPs ($\# \text{L}^{-1}$). It is well known that submicron particles are in high concentrations in comparison to supermicron particles; however, their mass is usually very low, in some cases at or below the detection limit of the analytical technique.

Technical comments

Line 10: should it be tropical instead of topical(?)

A: Fixed.

Line 35: It is encouraged that the author cites the original literature along with the newer cross-references. e.g. Kanji et al. (2017) cite other older references.

A: The text was modified to include references to the original literature as follows. Lines 34-38: “Murray et al. (2012) and Ladino et al. (2013) have suggested that contact freezing and immersion freezing are the most efficient mechanisms leading to ice nucleation in clouds; however, the atmospheric relevance of contact freezing is still under debate (Hobbs and Atkinson, 1976; Ansmann et al., 2005; Cui et al., 2006; Phillips et al., 2007; Seifert et al., 2011; Kanji et al., 2017).”

Line 41: Using the word “most important” may not be a good idea.

A: As suggested by the reviewer, “the most important” was replaced by “...has been recognized as a very important INP...”.

Line 110: It would be nice if “wet” is quantified in terms of relative humidity if the measurement is available.

A: The original manuscript (line 133) mentioned that the ambient RH during the field campaign was typically above 67%. Nevertheless, the RH was added to this sentence for clarity and it now reads. Line 114: “and only wet aerosol particles were sampled (mean RH=69 %)”.

Line 115: Figure 2 shows three types of time series for 3 different cut off of the particles sizes. Since the instruments used employ different principles of measurements, it would be appropriate to explain the instrument principle in brief.

A: The following text was added. Lines 122-127. “In the CPC, the size of the aerosol particles is increased in a heated saturator/cooled condenser system prior to their detection. The particles grown are directed towards a laser beam and the dispersed light is collected by a photodetector that convert it to particle concentration. Similar to the CPC, aerosol particles in the LasAir are counted by passing them through a laser beam (without any prior treatment). Based on the pulses (or voltage) and their amplitude the dispersed light by the particles is then converted to particle concentration and size.”

Line 235: It should be Fig. 3 instead of Figure 3.

A: Fixed.

Reviewer #4

This manuscript presents the properties of ambient particles and INPs in the tropical latitudes using several instruments and sampling techniques, in order to provide a comprehensive analysis of the chemistry and biological properties of the INPs. Characterizing INP concentration around the world and understanding the contribution of bioaerosols are important, and therefore I find this study a valuable contribution, especially since there is little data from tropical latitudes.

The introduction part is good and the manuscript is well written. However, at some parts of the manuscript I felt the need for more details, as in the ice nucleation part (see below). Also, I do not think that the title reflects the findings well. I think it confuses the reader to think there is some new evidence on biological INPs, however, it was not proved that the bioaerosols are responsible for the ice nucleation activity that was detected, and I personally still not convinced. Thus, I would recommend changing the title to a more suitable one, or provide the proof for the importance of the biological particles to the INP concentrations.

A: The requested details were added to the introduction and the title was modified to better reflect the results and conclusions. The revised title of the study is now: “Ice Nucleating Particles in a Coastal Tropical Site”

I think that the parts related to the freezing experiments and results, should be revised and clarified. There is not enough technical details about the method, such as the instrumentation or the temperature uncertainty, and what are the limitation of the analysis that was done.

A: More information regarding the ice nucleation experimental setup and uncertainties was added to the revised manuscript:

Lines 147-154: “The cold cell-microscope system used here is the same one used in previous studies (Mason et al., 2015a,b, 2016; DeMott et al., 2016; Si et al., 2018). The following steps encompass the analysis: i) The samples collected on glass cover slips were placed in the cold cell at room temperature, ii) The cold cell was isolated and kept at 0 °C, while humid air (RH>100 %) was injected into the cell to induce liquid droplets’ formation by water vapor condensation; iii) Dry air (N₂) was then injected into the cold cell to prevent the newly formed droplets from touching. This is a key step to minimize the probability of liquid droplets freezing by contact; and iv) Once droplets’ sizes and thermodynamic conditions were stable, the cold cell was closed.”

Lines 159-174: “The temperature at which each droplet froze was determined by analyzing the video from the CCD camera (XC-ST50, Sony) connected to the microscope and the data reported by the resistance temperature detector (RTD) located at the center of the cold cell with a ±0.2 °C uncertainty (Mason et al., 2015b). Homogeneous freezing experiments were performed on laboratory blanks exposed during the preparation of the MOUDI, while heterogeneous freezing experiments were run on ambient particles deposited on the glass cover slips (Figure S1). The [INP] was calculated using the following expression:

$$[INP_s(T)] = -\ln\left(\frac{N_u(T)}{N_o}\right) \cdot \left(\frac{A_{deposit}}{A_{DFTV}}\right) \cdot N_o \cdot f_{ne} \cdot f_{nu,0.25-0.10\text{ mm}} \cdot f_{nu,1\text{ mm}}, \quad (1)$$

where $N_u(T)$ is the number of unfrozen droplets at temperature T , N_o the total number of droplets, $A_{deposit}$ the total area of the aerosol deposit on the hydrophobic glass cover slip, A_{DFT} the area of the hydrophobic glass cover slip analyzed in the DFT experiments, V the total volume of air sampled, f_{ne} a correction factor to account for uncertainty associated with the number of nucleation events in each experiment, $f_{nu, 0.25-0.10\text{ mm}}$ and $f_{nu, 1\text{ mm}}$ a non-uniformity factor which corrects for aerosol deposit inhomogeneity at a scale of 0.25 - 0.10 mm, and 1 mm, respectively (Mason et al., 2015a). We refer the readers to Mason et al. (2015a) and Mason et al. (2015b) for more details of the MOUDI-DFT operational principle.”

Also - there are no error bars (or any error analysis) throughout the manuscript, which is very surprising, and must be done before resubmission.

A: The original manuscript included uncertainty bars in Figures 5 and 8; uncertainty bars have now been added to Figure 4, Figure 6, Figure 7, and Figure S4.

Specific comments:

Line 10: I guess should be “tropical” and not topical

A: Fixed.

Line 13: for -> from

A: Fixed.

Line 29: is -> was

A: Fixed.

Line 40: I suggest to detail how the concentration affect on water drop formation

A: Is the reviewer referring to the following statement: “Mineral dust has been recognized as the most important INP on a global scale due to their good ice nucleating abilities and their elevated concentrations in the troposphere”? If yes, we are referring to the INPs abilities of mineral dust and not to their CCN abilities. If there are more mineral dust particles, the likelihood that some of them can act as INPs is higher.

Line 88: present -> presents

A: Fixed.

Line 110: What is wet?

A: The original manuscript (line 133) mentioned that the ambient RH during the field campaign was typically above 67%. Nevertheless, the RH was added to this sentence for clarity and it now reads: Line 114: “and only wet aerosol particles were sampled (mean RH=69 %)”.

Line 235: if possible, please explain here how was the cold front determined, and send to the supplementary figures.

A: Figure 2A (Fig 3B in the original manuscript) shows the time series of the wind speed during the field campaign. Cold fronts are associated with significant changes in wind speed, wind

direction, and temperature. We use here changes in the wind speed data, which were more pronounced than in direction. Additionally, surface maps from NOAA were used to confirm the passage of the cold fronts as shown in Figure S3.

Line 250: Please explain why do you believe that there is no significant difference and that during front did not changed much. To me it seems that their slope is different and that the concentrations are also differ.

A: We agree with the reviewer. Both the slopes and concentrations are different, especially for submicron particles. The text was modified as follows. Lines 286-289: “As for the total aerosol concentration (Figure 2), the number size distributions of the aerosol particles larger than 300 nm were also impacted by the cold fronts. For example, the concentration of particles smaller than 5.0 μm was lower during the passage of the cold fronts (Figure S4).”

Line 265: I suggest rephrasing this sentence; especially the word important, it does not fit here.

A: The text was changed as follows. Lines 316-317: “for typical atmospheric concentrations of mineral dust, ice nucleation at these temperatures seems to be of secondary importance”.

Line 332: I suggest removing or rephrasing this paragraph; it is a speculation that is not accurate.

A: The sentence was modified to address the new results shown in Table 2.

Line 353: Can you detail what is the “uncertainties of analysis”

A: This was a grammatical mistake. Our intention was to acknowledge that a direct correlation between the MOUDI and biosampler data was not completely fair given that the different sampling time introduces uncertainty. The text was revised as follows. Lines 424-425: “This poor correlation can be in part due to the different sampling time of the MOUDI and the biosamplers”.

Line 451: fix reference of Chen et al. 2011.

A: We are sorry, but we could not find any mistake in this reference. Can the reviewer please let us known what is the problem here?

Chen, S.-C., Tsai, C.-J., Chen, H.-D., Huang, C.-Y., and Roam, G.-D.: The influence of relative humidity on nanoparticle concentration and particle mass distribution measurements by the MOUDI, Aerosol Sci. Technol., 45, 596–603, doi:10.1080/02786826.2010.551557, 2011.

Fig.2 – I suggest to write “entire” instead of “whole”

A: Changed.

Fig 2. Is seems to me that there is daily cycle of the aerosols? For example seen clearly in days 27-28.1.17.

A: We agree with the reviewer. Thanks for pointing this out. This is acknowledged in the revised manuscript. Lines 264-265: “From the CPC data shown in Figure 2A, there seems to be a daily cycle with most of the highest concentration taking place between 7 h and 12 h (local time)”

Figure S1. I would consider replacing the use of brackets in this caption. It was not clear to me at first what I see there.

A: The left), middle), and right) labels were replaced by A), B), and C) for clarity.

Figure S3. Why there are no error bars? That way it will be easier to understand if the two distribution are actually similar.

A: Uncertainty bars were added to three of the seven size distributions only for readability. When all seven size distribution have uncertainty bars, the figure is more difficult to understand.

Reviewer #5

“The Importance of Biological Particles to the Ice Nucleating Particle Concentrations in a Coastal tropical Site” by Ladino et al. describes efforts to characterize the INP population and biological particles at a tropical site. These data are valuable to the community due to a lack of data in such environments. My major comment is that more information on the INP measurement detection limits, blanks, and uncertainties is needed because the paper heavily relies on these data.

A: More information related to the experimental setup and its uncertainties was added to revised manuscript:

Lines 147-154: “The cold cell-microscope system used here is the same one used in previous studies (Mason et al., 2015a,b, 2016; DeMott et al., 2016; Si et al., 2018). The following steps encompass the analysis: i) The samples collected on glass cover slips were placed in the cold cell at room temperature, ii) The cold cell was isolated and kept at 0 °C, while humid air (RH>100 %) was injected into the cell to induce liquid droplets’ formation by water vapor condensation; iii) Dry air (N₂) was then injected into the cold cell to prevent the newly formed droplets from touching. This is a key step to minimize the probability of liquid droplets freezing by contact; and iv) Once droplets’ sizes and thermodynamic conditions were stable, the cold cell was closed.”

Lines 159-174: “The temperature at which each droplet froze was determined by analyzing the video from the CCD camera (XC-ST50, Sony) connected to the microscope and the data reported by the resistance temperature detector (RTD) located at the center of the cold cell with a ±0.2 °C uncertainty (Mason et al., 2015b). Homogeneous freezing experiments were performed on laboratory blanks exposed during the preparation of the MOUDI, while heterogeneous freezing experiments were run on ambient particles deposited on the glass cover slips (Figure S1). The [INP] was calculated using the following expression:

$$[INP_s(T)] = -\ln\left(\frac{N_u(T)}{N_o}\right) \cdot \left(\frac{A_{deposit}}{A_{DFT}V}\right) \cdot N_o \cdot f_{ne} \cdot f_{nu,0.25-0.10\text{ mm}} \cdot f_{nu,1\text{ mm}}, \quad (1)$$

where $N_u(T)$ is the number of unfrozen droplets at temperature T , N_o the total number of droplets, $A_{deposit}$ the total area of the aerosol deposit on the hydrophobic glass cover slip, A_{DFT} the area of the hydrophobic glass cover slip analyzed in the DFT experiments, V the total volume of air sampled, f_{ne} a correction factor to account for uncertainty associated with the number of nucleation events in each experiment, $f_{nu,0.25-0.10\text{ mm}}$ and $f_{nu,1\text{ mm}}$ a non-uniformity factor which corrects for aerosol deposit inhomogeneity at a scale of 0.25 - 0.10 mm, and 1 mm, respectively (Mason et al., 2015a). We refer the readers to Mason et al. (2015a) and Mason et al. (2015b) for more details of the MOUDI-DFT operational principle.”

I have several other concerns in the interpretation of results, described below. In general, the authors motivate the study by describing the need to characterize marine INP sources in the tropics (“Very few studies to sample INPs have been carried out in tropical latitudes, and there is a need to evaluate their availability to understand the potential role that marine aerosol may play in the hydrological cycle of tropical regions”), but I am not convinced the method deployed can measure [INP] for a remote region and I think the concentrations reported could not possibly be explained by marine aerosol.

A: As shown in Mason et al. (2016), this method has been successfully used in different remote regions, such as Alert (Canada), Labrador Sea, Ucluelet (Canada). Additionally, it is important to mention that this method has been correlated and validated with other ice nucleation instrumentation such as the Colorado State University-Continuous Flow Diffusion Chamber (CSU-CFDC), obtaining comparable results (DeMott et al. 2016). There are more than eight papers already published using the same technique used in this study. Additionally, the [INP] reported in this study (Figure 5) is on the same order (albeit slightly higher), than those reported in other coastal/marine locations using the same technique (Mason et al. 2015a).

Finally, as illustrated in Figure 8 the chemical analysis performed by two different analytical techniques (i.e., HPLC and XRF) shows the presence of Na and Cl in significant concentrations, confirming that the air masses sampled were clearly maritime.

Furthermore, additional analysis carried out on samples collected at the same site in July (not relevant for the manuscript under review, and not yet published), is presented here to address the reviewer’s concern about the maritime origin. The Table below shows particle chemical composition sampled in Sisal under the influence of Saharan dust (July 2018), with a dominant signature in Si, Fe, and Al, instead of the Na and Cl dominant in the wintertime samples discussed in the present study.

Table R1. Comparison of the aerosol chemical composition in Sisal during winter 2017 and summer 2018.

Element	XRF concentration in Winter 2017 (ng m ⁻³)	XRF concentration in Summer 2018 (ng m ⁻³)
Na	1.83	0.38
Cl	5.49	0.23
Al	0.11	1.00
Si	0.22	1.72
Fe	0.21	0.96

The new analysis performed (Table 2 in the revised manuscript) suggests that mineral dust particles could be responsible for INPs between -20 °C and -30 °C. However, this is not the case for INPs measured at -15 °C. This is now discussed in the revised manuscript.

Bibliography cited in this reply:

DeMott, P. J., Hill, T. C., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., et al.: Sea spray aerosol as a unique source of ice nucleating particles, *P. Natl. Acad. Sci.*, 113, 5797–5803, doi:10.1073/pnas.1514034112, 2016.

Mason, R., Chou, C., McCluskey, C., Levin, E., Schiller, C., Hill, T., Huffman, J., DeMott, P., and Bertram, A.: The micro-orifice uniform deposit impactor–droplet freezing technique (MOUDI-DFT) for measuring concentrations of ice nucleating particles as a function of size:

improvements and initial validation, *Atmos. Meas. Tech.*, **8**, 2449–2462, doi:10.5194/amt-8-2449-2015, 2015a.

Mason, R., Si, M., Chou, C., Irish, V., Dickie, R., Elizondo, P., Wong, R., Brintnell, M., Elsassner, M., Lassar, W., et al.: Size-resolved measurements of ice-nucleating particles at six locations in North America and one in Europe, *Atmos. Chem. Phys.*, **16**, 1637–1651, doi:10.5194/acp-16-1637-2016, 2016.

Overall, I think there are a few things that need to be clearly stated and supported consistently throughout: 1) What were the [INP] and their variability (and their detection limits)?

A: This information is now provided in the revised Figure 4 and Figure S5. The following text was added to Figure captions: “The upper and lower detection limits of the MOUDI-DFT are 30 L⁻¹ and 0.01 L⁻¹, respectively.”

2) What size range corresponded to the highest [INP]?

A: This information is now shown in new Figure 4. The following text was added. Lines 311-313: “The “high” [INP] found at -15 °C can be explained in part by the very efficient INPs shown in Figure S6, with sizes ranging from 1.0 μm to 1.8 μm. However, it is important to note that particles with diameters between 1.8 μm and 10 μm also contribute to the total [INP] at warm temperatures.”

3) what meteorological conditions, air mass histories (back trajectory and PCR results) corresponded to the highest [INP]?

A: Figures 5 and 7 indicate that the cold air masses behind the cold fronts show higher [INP].

4) What is the hypothesized origin of these very high [INP] and biological particles? If the authors can build up the results discussions around some clear points, I think it will be easier to follow along.

A: Figure 4, 6, and 7 suggests that the more efficient INPs are those supermicron in size. Additionally, based on the results summarized in Tables 2 and 3, and given that at -15 °C mineral dust is not the likely source of the measured INPs, we hypothesize that they are biological (both continental and marine). However, given that the [INP] under the influence of cold fronts is higher, it may be more likely that the efficient biological INPs are of marine origin.

The following text was added. Lines 482-487: “Based on the large [INPs] above -15 °C, the supermicron size of 90 % of the INPs, the presence of marine biological particles in the cold air masses those of which showed the highest [INP], in addition to the poor correlation shown by the mineral dust tracers with the [INP], we hypothesize that the likely source of the INPs measured at high temperatures in Sisal are biological particles. Therefore, our results suggest that continental and maritime biological particles could play an important role in ice cloud formation and precipitation development in the Yucatan peninsula.”

Comments:

Abstract: Should mention the freezing mode and INP temperature range measured during this study in abstract.

A: This is now stated in the revised version. Lines 12-13: “Aerosol particles sampled in Sisal are shown to be very efficient INPs in the immersion freezing mode, with onset freezing temperatures...”

L18 – I think a better way to say this is similar to how it was stated in the results section, something like “The high concentrations of INPs at warmer ice nucleation temperatures ($T > -15^{\circ}\text{C}$) and the supermicron size of the INPs suggest that biological particles may have been a significant contributor to the INP population in Sisal during this study”.

A: We thank the reviewer for the suggestion and the text was revised accordingly.

L55: The modeling studies listed determined specific regions where oceanic sources dominated the INP population due to an absence of other types, like mineral dust.

A: The sentence was modified as follows for clarity. Lines 56-60: “Marine organic matter, likely of biological origin, has been suggested to be an important oceanic source of INPs in the southern oceans, north Atlantic, and north Pacific (Burrows et al., 2013; Yun and Penner, 2013; Wilson et al., 2015; Vergara-Temprado et al., 2017); however, the maritime source suggestion was made with little or no data from tropical latitudes”.

L58: Bigg (1973) was the first to report such a study (his data are used in Schnell and Vali, 1975).”

A: Thank you for pointing this out. The citation to the Bigg (1973) study was added to the revised manuscript.

L67 – the range may also be from different species, right?

A: You are correct. However, this is a very general statement that aims to introduce the large [INP] range in marine environments. We do not see the need to modify the statement.

L142 – please provide information on the measurement detection limit and how measurement uncertainty was determined. Where there blanks collected and how were these accounted for?

A: See the first answer to reviewer #5 above. Homogeneous freezing experiments were performed on blanks. The blanks refer to clean glass covered slips exposed while mounting and preparing the MOUDI in the lab during the field campaign. The figure R2 included below was added to the supplementary material and it shows examples of activation scans for blanks and ambient particles.

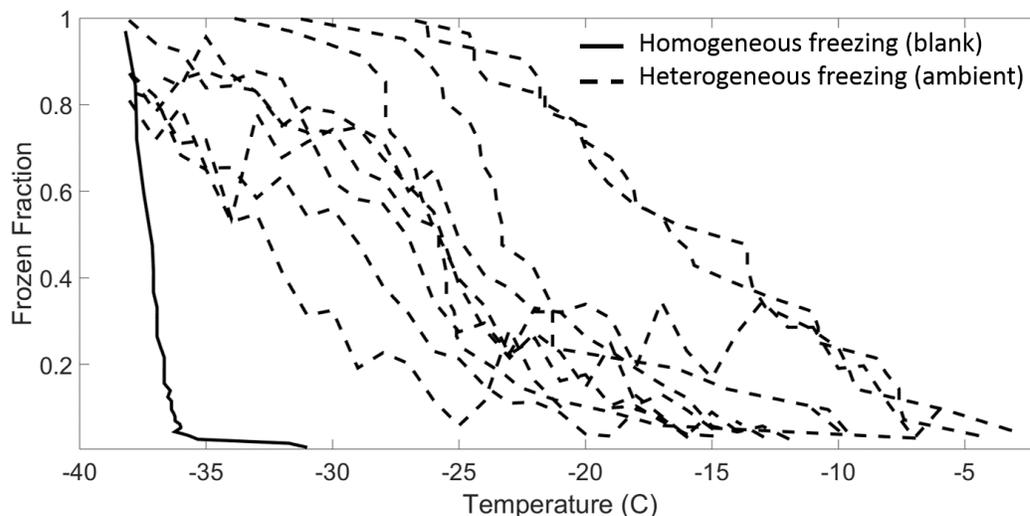


Figure R2. Example of activation curves for the blanks (homogeneous freezing) and ambient particles (heterogeneous freezing).

Fig S3- Why are there no particles larger than 1 micron at a coastal site? Is this consistent with other studies? Are the y axis units correct?

A: We thank the reviewer for pointing out the issues on Figure S3 (now Figure S4). The LasAir reports the concentrations in # m⁻³ and # ft⁻³ and we were using # ft⁻³ instead of # m⁻³ in the original manuscript. The values were converted from # ft⁻³ to # m⁻³ in the revised manuscript. Additionally, we found out that we had used the incorrect size bins. In the original manuscript we inadvertently used the size bins corresponding to the recently acquired OPC (LasAir III 310B from 0.3 μm to 10 μm) instead of those of the old one used for this study (LasAir II 310A from 0.3 μm to 25 μm). These important issues are now fixed in the revised manuscript, and we thank the reviewer for bringing them into our attention.

L250 – Did you compare particle composition between the cold front/marine air mass periods and the other periods? These back trajectories shown in Figure S1 suggest that the air masses actually originated from the US Central Plains. So, you would expect a mixture of aerosol composition I think.

A: Figure R3 included below was added to the revised manuscript with the following text. Lines 289-295: “As shown in Figure 3 (and Figure S5), the XRF analysis indicates that although there are small differences in the bulk chemical composition of the aerosol particles in the presence/absence of cold fronts, in general, they are rather comparable. Note, however, that this is not a completely fair comparison given that sampling time for the chemical analysis was 48 h, while sampling time for determining the influence of the cold air masses was on the order of 36 h. Therefore, the periods denoted as cold fronts contain aerosol particles that may not technically belong to this category.”

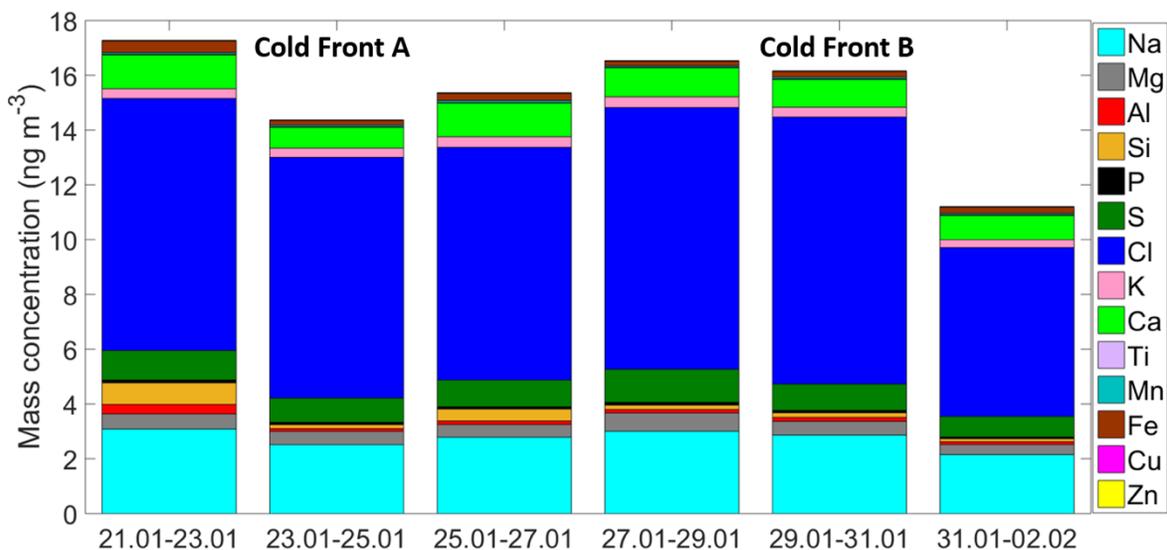


Figure R3. Time series of the ambient aerosol mass concentration and bulk chemical composition as measured by the XRF. Each sample was collected for 48 h starting at 12:00 h local time.

The following sentence was also added to acknowledge this. Lines 366-369: “Also, it is important to note that although the cold air masses that reached Sisal behind the cold fronts had crossed the GoM, the aerosol particles found in them are likely a mixture of particles originated in the US Central Plains and the GoM (Figs. S2B-C and S5). ”

Fig 4 – It would be best to show the data from this work as points versus a shaded region so that the variability in [INP] is fully illustrated/reported. Are these samples background corrected? Are the [INP] for all the stages combined or each individual stage? For temperatures lower than $\sim -25^{\circ}\text{C}$, the “bluish” region flat-lines at about 30 L^{-1} – is that the upper detection limit of the INP measurement? Same with lower detection limit. This figure suggests to me that the range of detection of this method is from 0.1 L^{-1} to 30 L^{-1} . Is it possible for this method to observe the concentrations reported for remote marine environments (dark blue shade, DeMott et al. (2016))? These detection limits should be noted in the methods and in the figure caption.

A: We agree with the reviewer’s suggestion and the figure was completely modified as shown below. This new figure shows the average [INP] as a function of MOUDI stage and also the combined concentration (as grey triangles). The large amount of data from Kanji et al. (2017) were removed in the revised figure and only include for comparison, the coastal/oceanic data from DeMott et al. (2016), Welti et al. (2018) and Irish et al. (2019).

The upper and lower detection limits of the MOUDI-DFT are 30 L^{-1} and 0.01 L^{-1} , respectively. This information was added to the revised Figure caption. Regarding background correction, although blanks were collected a correction was not necessary. As shown in Figure S1, in the blanks, ice formation was not observed above -30°C .

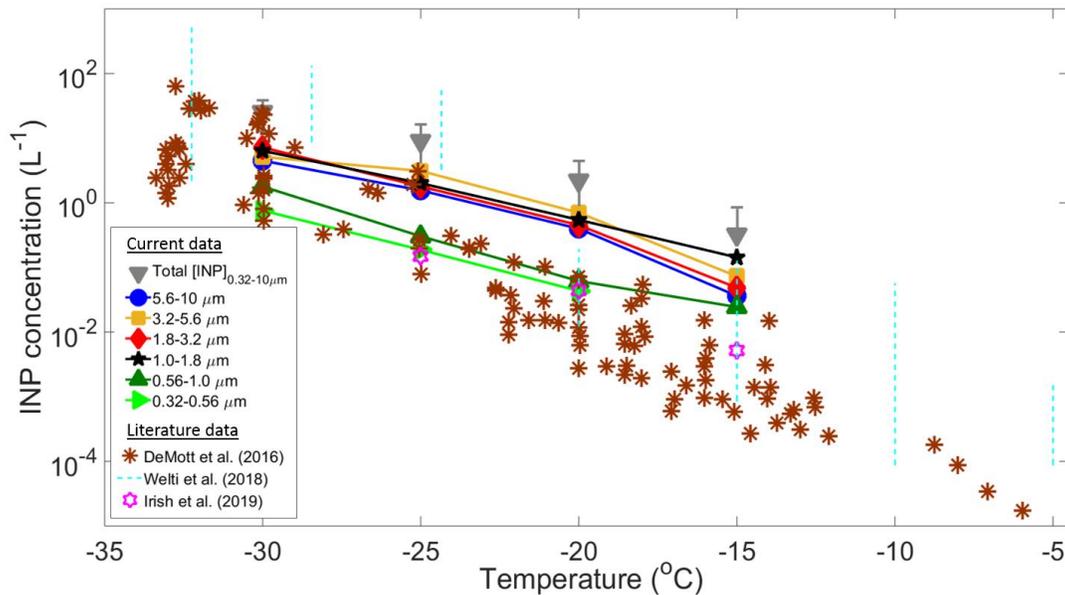


Figure R1. Summary of the INP concentrations as a function of temperature and particle size (solid symbols). Mean [INP] are represented by the grey triangles, whereas the brown asterisks, light blue dotted lines, and purple stars are literature data from DeMott et al. (2016), Welti et al. (2018), and Irish et al. (2019), respectively. The upper and lower detection limits of the MOUDI-DFT are 30 L⁻¹ and 0.01 L⁻¹, respectively.

L260 –Should also note that the [INP] reported here are up to 3-4 orders of magnitude higher than [INP] reported for marine boundary layer measurements reported by DeMott et al., 2016.

A: This is now acknowledged in the text, as follows. Lines 304-307: “At -15 °C the [INP] measured in Sisal are in relatively good agreement with those found at Cabo Verde (Welti et al., 2018) but are one to two orders of magnitude higher than the values reported by Irish et al. (2019) from the Arctic boundary layer and by DeMott et al. (2016) from sea spray laboratory generated particles and ambient marine boundary layer particles”

L270 – “The Sisal data corresponds to particle diameters ranging between 0.32 μm and 10 μm where 16 out of the 29 samples fulfilled the size criteria.” – please clarify what is meant by this?

A: This sentence is meant to indicate that out of the 29 samples analyzed for this study, we have information in the size range that goes from 0.32 μm to 10 μm in only 16 of them. Each “sample” consists of 6 glass covered slips, which are not always usable. For example, they can break during the analysis, or the analysis itself can fail, and therefore no information can be recorded. Therefore, we have a full dataset (from all 6 covered slips) in 16 out of the 29 samples. The sentence was modified as follows:

Lines 322-323: “The Sisal data corresponds to particle diameters ranging between 0.32 μm and 10 μm; full information in all size stages was obtained in 16 out of the 29 samples analyzed”

L283 – what time of year where Rosinski’s measurements made?

A: The following text was added. Line 339: “between July 20 and August 30, 1986”

Fig 6 – Should there be standard deviation bars on these? Also, if one were to use Figure 6 and Figure S3 to determine a number fraction (which should be done as an analysis), the number fractions are bogus. Are the units of Figure S3 correct (maybe they should be per cubic centimeter)? How do you have higher [INP] than total particle counts in the same size bin?

A: The corresponding standard deviations were added to Figure 6. The units in Figure S3 were in # ft⁻³ and as explained above, they were converted to # m⁻³ in the revised manuscript. The inconsistency was caused by the error in units.

The fraction of particles acting as INP is now shown in Figure 7. Lines 372-380: “To confirm the size dependence and the importance of supermicron particles to the [INP] in Sisal, the fraction of particles acting as INPs was calculated combining the DFT and LasAir data (Fig. 7). The [INP] was normalized for four size bins (i.e., 0.3 μm-0.5 μm, 0.5 μm-1.0 μm, 1.0 μm-5.0 μm, and 5.0 μm-10.0 μm). As expected (from Figs. 4 and 6), the fraction of particles acting as INPs increases with increasing particle size and with decreasing temperature. This trend is in agreement with the results shown by Si et al. (2018), with the present results being higher. Figure 7 also shows that the fraction of aerosol particles acting as INP is higher when influenced by the cold fronts (black symbols), especially for particles ranging between 1.0 μm and 5.0 μm.”

Fig 7/L309 – Are these results for the entire study or a specific period? Please add this detail to figure caption and text. If the entire study, why not look at individual events? They were 48 hours sample, so perhaps show a timeline? Is it not possible to look at carbon or oxygen with this method?

A: The results are for the whole period. This was added to the figure caption and text. Additionally, Figure 4 and S5 show the timeline of the XRF results.

L328 – show the timeline in the supplemental to support this statement?

A: This sentence was deleted and new information was added (i.e., Figure 4 and Figure S5)

Figure S4 – “Daily profile” or is this the average of two days of data (i.e., two points averaged for each time bin)?

A: This is a daily profile. A sample was collected every two hours for 24h to get the information reported in Figure S4 (Now Fig. S7).

Fig 8 – why do only some [INP] points have horizontal lines? The y axis on the top two panels have errors for the lower limit label. What are the measurement uncertainties for [INP] and bacteria/fungi?

A: We added the horizontal lines to some of the [INP]_{-15C} to illustrate the time span but as the time is constant we did not add it to every single data point. It is not possible to add horizontal uncertainty bars to the bacteria and fungi data as the sampling time was only 5 minutes, and therefore, the size of the uncertainty bar is much smaller than the size of the symbols (black stars and red diamonds). Example uncertainty bars were added to the [INP] and fungi/bacteria concentrations.

L365 – were offshore chlorophyll a concentrations elevated during this study?

A: Unfortunately, we did not have the means to measure chlorophyll a in the present study.

L366 – I think it's great to show the utility of this method for showing the air mass history (i.e., terrestrial versus marine). I suggest pulling this forward in the introduction, as this a unique approach to identify air mass origin (e.g., In this study, we use PCR to confirm air mass history and its influence on [INP]) and also reference any other papers that have attempted this (if applicable).

A: We don't get the point of the reviewer here. We are not trying to confirm the air mass history by PCR analysis (i.e., identification of the bacteria and fungi species). This is not claimed in the manuscript and it is not our intention to acknowledge this. Our purpose was: i) to confirm the presence of biological particles in the air masses sampled, ii) if present, to linked them with potential sources based on the literature, and iii) to see if the viable microorganisms differ when cold air masses arrived in Sisal.

L387 – The dates of the study should be specified here (for those who read only the conclusions..)

A: Added.

L390 – Should report the range of [INP] for a given temperature

A: The text was modified as follows. Lines 464-467: "The results show that the INPs concentrations in Sisal are comparable ($0.33\pm 0.53\text{ L}^{-1}$, $2.20\pm 2.26\text{ L}^{-1}$, and $8.96\pm 7.38\text{ L}^{-1}$ at $-15\text{ }^{\circ}\text{C}$, $-20\text{ }^{\circ}\text{C}$, $-25\text{ }^{\circ}\text{C}$, respectively) and in specific cases even higher than at other locations studied using the same INP counter type, especially under the influence of cold fronts."

L390 – also similar to [INP] measured from U.S. Central Plains (harvesting aerosol), as you mention in the text. I think it should also be clear that the [INP] are high for a marine environment (i.e., comparison to DeMott et al., 2016 marine measurements).

A: This sentence was modified in accordance with the new Figure 4 (Figure R1). See answer for L260 above.

L406 - Could you comment on the representativeness of these measurements for modeling efforts? I.e., would you expect these [INP] to change for different seasons based on Rosinski's work? What size bins would you expect to reach cloud level and therefore what [INP]?

A: The following text was added to address the reviewer's concerns. Lines 492-497: "The present results are important for the development of new parametrizations to be incorporated in climate models given that the currently available parametrizations contain little or no data from tropical latitudes. However, further similar studies are needed given that the [INP] may vary seasonally. Especially, the arrival of mineral dust particles to the GoM and the Caribbean region from Africa in July-August are expected to impact the [INP] and therefore, ice cloud formation, as shown by Rosinski et al., (1988) and DeMott et al., (2003)".

Our own results also confirm the arrival of the Saharan dust to Sisal as shown in Table R1. The impact of these particles on the [INP] will be shown in a follow-up study.

Technical comments:

L2 – "are referred to as ice nucleating particles (INP)." Should be: "are referred to as ice nucleating particles (INPs)."

A: Fixed.

L3 – “mid- and high-latitude oceans” – I think there is general consensus that bubble bursting at the ocean surface (regardless of latitude) is a source of aerosol.

A: We agree with the reviewer; however, we are not referring to bubble bursting as a source of aerosol particles. We are referring to the recent studies conducted in mid- and high-latitudes that showed that marine aerosol have the potential to act as INPs. This is more fully explained in the introduction. Lines 61-65: “Important efforts were made during the 1950-70’s to understand the role of the oceans in ice cloud formation (Bigg, 1973; Schnell and Vali, 1975; Schnell, 1975, 1977, 1982; Rosinski et al., 1987, 1988). There is recent new and robust evidence that biological material from the marine environment could act as efficient INPs (Knopf et al., 2011; Wilson et al., 2015; Mason et al., 2015b; DeMott et al., 2016; Ladino et al., 2016; McCluskey et al., 2017; Irish et al., 2017; Welti et al., 2018).”

L11 – may be helpful to add the latitude here since your reference “similar latitudes”

A: This was added. Lines 11-12 : “...and one of the few datasets currently available at such latitudes (i.e., 21 °N).”

L18 – “Biological particles were likely found to be very important” should be “Biological particles were found to be likely important”

A: The sentence was modified following your suggestion above.

L19 – “A variety of bacteria and fungi were identified.” – identified as what?

A: This sentence was removed

L20 – “Although the majority are of terrestrial origin, some of them are clearly oceanic.” – majority of what? What is “them”?

A: This sentence was removed

L95 – fix lat/lon format

A: Fixed.

Fig 1 – would be beneficial to add a scale bar to this photos

A: A scale bar was added.

L138 – is stage one 0.18 micron or 10 micron?

A: This is now clarified as follows. Lines 156-159: “Stage one (cut-size of 10.0 µm) was not taken into account given that the aerosol concentration on the glass substrates was typically very low, whereas in stage eight (cut-size of 0.18 µm) the number concentration of particles deposited on the glass substrates was so high that inhibited the proper formation of water drops.”

L276 – Are these error bars a standard deviation? Please define in figure caption

A: Yes. Added to figure caption.

Table 3 – does this source correspond to all of the Genus listed?

A: Yes. It is not possible to quantitatively assign a source by Genus.