

## Responses to Reviewer's and Editor Comments

**We would like to thank the Reviewers and the Editor for their valuable additional comments and suggestions. Specific answers and revisions to the text and figures related to each comment are given below in red bold text.**

### Editor

- Line 3, page 1: Why only "mid- and high-latitude" oceans? There is no reason why other oceans shouldn't release material that can act as INP. Suggest to delete "mid- and high-latitude"

**A: Deleted.**

- Line 5, page 1: Delete "the" before oceans.

**A: Deleted.**

- Line 37, page 2: Can a reference from 1976 still be part of a current debate? Suggest to reformulate this sentence.

**A: The sentence was modified as follows (Lines 36-37): "however, the atmospheric relevance of contact freezing is still unclear given the contradictory results".**

- Line 99, page 4 and throughout the manuscript: The geographic abbreviations (N, W) should not be in italics (remove the Latex mathmode).

**A: Fixed.**

- Line 109, page 4: For completeness, add "(arithmetic mean  $\pm$  standard deviation)"

**A: Added.**

- Line 129, page 5: Please mention that these are "optical diameters". Is the cut-size the upper, lower or mid-bin size? I guess the last bin counts all particles larger than 25 micron?

**A: "Optical diameter" was added.**

- Line 137, page 5: Please precise the size ranges of the different stages. I guess the first one is ">10 micrometer", the second one "10-5.6 micrometer", etc.? See also line 156.

**A: The following text was added to the revised manuscript (Lines 139-140): “The particle size range for each MOUDI stage are given in Table S1”. Table S1 was added to the Supplementary Material and the size range was added to Line 156.**

**Table S1.** Particle size range for each MOUDI stage.

| <b>MOUDI stage</b> | <b>Size range (<math>\mu\text{m}</math>)</b> |
|--------------------|--|
| <b>1</b>           | <b>&gt;10.0</b>                              |
| <b>2</b>           | <b>5.6 – 10.0</b>                            |
| <b>3</b>           | <b>3.2 – 5.6</b>                             |
| <b>4</b>           | <b>1.8 – 3.2</b>                             |
| <b>5</b>           | <b>1.0 – 1.8</b>                             |
| <b>6</b>           | <b>0.56 – 1.0</b>                            |
| <b>7</b>           | <b>0.32 – 0.56</b>                           |
| <b>8</b>           | <b>0.18 – 0.32</b>                           |

- Equation 1, line 166 and later in the manuscript: The subscripts in the equation or of variables in general should not be in italics. Add “\rm” in the parenthesis (e.g.  $A_{\text{deposit}}$ ). Instead of “ln” use “\ln” since this is a function.

**A: Fixed.**

- Line 254, 255 and also later in the manuscript: The capitalization of the section headings suddenly changes compared to previous headings ... Please harmonize.

**A: Fixed.**

- Line 258, page 8 and throughout the manuscript: Within the sentence the word figure should be “Fig.” while “Figure” when it is at the beginning of the sentence. Please have a look at [https://www.atmospheric-chemistry-and-physics.net/for\\_authors/manuscript\\_preparation.html](https://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html) and carefully fulfil the manuscript preparation requirements.

**A: Fixed here and along the manuscript.**

- Line 273, page 9: “than” -> “that”, add “the” before “cold”

**A: Added.**

- Line 276, page 9: “Hysplit” -> “HYSPLIT”, move “from the measurement site” to the front “Back-trajectories from the measurement site ...”

**A: Fixed.**

- Line 296, page 9: I suggest to remove the parenthesis “([INP])” from the heading title, it should be defined within the text (as has been done before).

**A: Deleted.**

- Line 400, page 12: O’Dowd

**A: Fixed.**

- Line 415, page 13: Good correlation with what exactly? INP concentration?

**A: The sentence was modified as follows (Lines 434-435): “The bacteria and fungi concentrations showed a relatively good correlation between each other.”**

- Line 460, page 14: Add “collected” or “sampled” before “around Sisal”.

**A: “collected” was added.**

- Line 466, page 14: Maybe better: “... using the same type of INP counter.” I would also start the issue of the influence of the cold fronts in a new sentence.

**A: The sentence text was modified as follows (Lines 485-487): “at other locations studied using the same type of INP counter. The higher INP concentrations were observed especially under the influence of cold fronts.”**

- Line 468, “than” -> “that”

**A: Fixed.**

- Line 473: Better: “However, the concentrations of Al, Si, ...”

**A: Changed.**

- Line 476: “magnesium” -> “Mg” (to be consistent)

**A: Fixed.**

- Page 14 & 15: Large parts of the conclusions, especially the last three paragraphs, are more a discussion than a precisely formulated conclusion and should be moved to Sect. 3 (Results and Discussion).

**A: The following text was moved from the “Conclusions” to the “Results and discussion section” to improve the readability of the manuscript:**

**Lines 413-417: “From the correlation of the [INP] and the aerosol chemical composition at -15 °C, Mg was the only element showing a correlation that is statistically significant at the 95 % confidence interval ( $p < 0.05$ ). Although Mg can be found in mineral dust particles in low percentages, it can also be found in marine environments linked to sea spray aerosol (e.g., Savoie and Prospero (1980); Andreae (1982); Casillas-Ituarte et al. (2010)).”**

**Lines 429-430: “As stated by Islebe et al. (2015) both bacteria and fungi need to be properly documented in the peninsula and the GoM to fully understand their regional importance.”**

**Lines 423-426: “Efficient INPs such as those measured in Sisal could be very important for cloud glaciation. Additionally, they can trigger ice multiplication or secondary ice formation at such high temperatures via the Hallett-Mossop mechanism (Hallett and Mossop, 1974; Field et al., 2017) and impact precipitation formation.”**

- A statement on the data availability is missing (see [https://www.atmospheric-chemistry-and-physics.net/about/data\\_policy.html](https://www.atmospheric-chemistry-and-physics.net/about/data_policy.html)).

**A: The following text was added (Line 517): “Data availability. Data are available upon request to the corresponding author.”**

- Table 1: Also add the acronyms for HLPC and XRF.

**A: Added.**

- Table 2: Please add a column showing how many sample points are being compared here.

**A: Given that the number of points is the same for every temperature we think that adding a column is not necessary. The number of points is now added to the Table’s caption.**

### **Reviewer #1:**

The authors have improved their manuscript according to recommendations, including the representation of aerosol concentrations in terms of geometric mean and multiplicative standard deviation (i.e. lines 260-264).

**A: We thank the reviewer for its positive feedback.**

For readers not familiar with the concept, it would be helpful to slightly re-write this sentence and add the term "multiplicative standard deviation", for example in this way: "There is a large diurnal variability for the aerosol particle concentration measured by the CPC (particles > 30 nm, Fig. 2B) and the LasAir (particles >300 nm, Fig. 2C). Assuming log-normal distributions, the geometric mean concentration and multiplicative standard deviation (c.f. Limpert et al., 2001) for the entire sampling period was  $758.51 \times / 1.76 \text{ cm}^{-3}$  and  $1.00 \times / 1.37 \text{ cm}^{-3}$ , respectively." In addition, authors may consider to present [INP] in the Conclusion section also in terms of geometric mean and multiplicative standard deviation. As it is now, the value of the standard deviation (assuming normal distribution) is larger than the mean (at  $-15^\circ\text{C}$  and  $-20^\circ\text{C}$ ), suggesting there were instances with negative number concentrations of INP, which does not make sense.

**A: The suggested text was added to the revised manuscript. Additionally, the following text was added to the conclusions (Lines 483-485): "(geometric mean and multiplicative standard deviation of  $0.44 \times / 1.77 \text{ L}^{-1}$ ,  $1.73 \times / 2.56 \text{ L}^{-1}$ , and  $6.20 \times / 2.65 \text{ L}^{-1}$  at  $-15^\circ\text{C}$ ,  $-20^\circ\text{C}$ , and  $-25^\circ\text{C}$ , respectively)".**

line 257: change "aire" to ""air"

**A: Fixed.**

line 263: change "than" to "that"

**A: Fixed (We think the reviewer refers to line 273 instead of line 263).**

#### **Reviewer #4:**

I do not recommend on this study to be published in acp, mainly because I do not see new findings that add scientific progress. The conclusions are still relying on other literature, more than on findings from this study. I think the technical part of this study was done well, and that the resulting data is worth publishing, especially due to the fact that there is not much data on INPs from this region, however, from a manuscript that is submitted to acp, it is expected that the data analysis and conclusions will be more comprehensive and innovative, as well as to fulfil its scientific potential.

**A: We disagree with the reviewer recommendation, especially because we have properly addressed each of the 20 points listed in his/her original review. In the revised manuscript we added new information to support our conclusions: 1) correlation of the [INP] with the bulk aerosol chemical composition (Fig. 3 and S5), 2) correlation of the [INP] with the total aerosol concentration and particle size (Fig. 7). These are novel measurements for this tropical site and this is one of the few studies reporting the [INP] as a function of particle size covering such a large size range (0.18 to  $10 \mu\text{m}$ ). In summary, the present study not only reports the [INP], but it also provides information regarding the chemical, physical, and biological characteristics of the collected aerosol in addition to the meteorological variables. The reviewer evaluation is in contrast with the**

**other two reviewers who evaluated the revised manuscript from “Good” to “Excellent” in all three categories.**

**Reviewer #5:**

Ladino et al., have thoroughly satisfied my comments and their manuscript contributes a valuable analysis and dataset for the ice nucleation community. I think the paper should be accepted for publication after addressing the following mostly minor comments.

**A: We thank the reviewer for its positive feedback.**

L57: “important oceanic sources of INPs” suggest change to “important oceanic source of INPs”

**A: Changed.**

L105/figure1: could a label be added for Sisal for those not as familiar with the geography of this region?

**A: The location of the Center of Sisal was added to Fig. 1.**

L124: “... photodetector that convert it...” change to “...photodetector that converts it...”

**A: Changed.**

L170: can the authors expand on the correction factor ( $f_{ne}$ )? Is this determine each run or for each study? What range of factors are used? Is this a passion counting statistic uncertainty?

**A: From Demott et al. 2017. “ $F_{ne}$  is a correction factor to account for the statistical uncertainty that results when only a limited number of nucleation events are observed.  $f_{ne}$  was calculated following the approach given in Koop et al. (1997) using a 95 % confidence interval.”  $F_{ne}$  is based on Poisson counting statistics and it was calculated for each run.**

L174: I think the detection limits of the MOUDI-DFT need to be included in the methods description also (rather than just the Figure 4 caption). This is an important distinction from other measurements shown on Figure 4. That is, there may be [INP] present at Sisal that are below or above the instrument’s detection limit, so it’s not a very good direct comparison at all temperatures. E.g., DeMott et al., reported measurements less than 0.0001 L-1, but those are not measurable by the MOUDI-DFT.

**A: The detection limits of the MOUDI-DFT were added to the Methods (Lines 175-176).**

L257: typo: “different aire mass”

**A: Fixed.**

L263: I think there may be a formatting issue here for the mean number concentrations

**A: This is the multiplicative standard deviation format used by Limpert et al. (2001) when using a log-normal distribution. The text was modified following Reviewer’s #1 suggestion.**

L272: Any precipitation from these frontal passages? This may be another explanation for lower concentrations and suppressed diurnal cycle.

**A: We thank the reviewer for bringing this up. The following text was added (Lines 276-278): “During the passage of cold front A, precipitation events were not observed which was not the case for cold front B. This could partially explain the lower aerosol concentration observed during the passage of the cold front B in comparison to cold front A”.**

L276: This is typically written in all caps (HYSPLIT)

**A: Fixed.**

L289: “..during the passage of the cold fronts” should be “...during the passage of Cold Front B” I think.

**A: The reviewer is right. This was fixed.**

Figure 3: Should the start time be 00:00 h local time? The xlabel suggests that the start time was midnight?

**A: The starting time for the XRF samples was always at 12:00 h (local time). In this case the Figure caption is correct. Given the mismatch between the length of the cold fronts and the XRF sampling time, the addition of the Cold front A and B labels on Figure 3 could confuse the readers. Therefore, the “Cold front A” and “Cold front B” labels were changed by “A” and “B” indicating that these samples were partially influenced by the passage of the cold fronts A and B. The following text was added to the Figure’s caption: “A and B indicate that those samples were partially influenced by the passage of the Cold front A and the Cold front B, respectively”.**

L290: sentence contains grammatical error?

**A: The sentence was modified as follows (Lines 296-298): “the XRF analysis indicates that although there are small differences in the bulk chemical composition of the**

**aerosol particles, the overall composition is generally comparable in the presence or absence of cold fronts.”**

Figure S4 – Are these 24 hour averages? Perhaps add this detail to the figure caption.

**A: Yes. This was added to the figure caption.**

L292: suggest change to: “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 front air masses on INP populations was on the order of 36 hours. Therefore, the periods denoted as cold fronts contain aerosol particles that may not technically correspond to cold front air masses.” (or something like this)

**A: Thank you for the suggestions. This was added to the revised version (Lines 298-333).**

Figure 4: “Summary of INP concentrations” suggest change to “Summary of average INP concentrations”; what are the vertical lines for the grey points, 1 standard deviation? As a suggestion to highlight the number of samples and work that was included in this project, I would pull each of the values from Figure S6 into Figure 4 as lighter colors. There is a lot of data here, and it could be lost if all just placed in the supplemental.

**A: The figure caption was changed as suggested. However, we do not feel comfortable with the second suggestion (i.e., to add the data from Fig. S6 into Fig. 4). Adding the data from Fig. S6 into Fig. 4 will make Fig. 4 very messy and difficult to follow. Additionally, the data from Fig S6. will not add additional information to Fig. 4.**

Table 1 – could move this to supplemental I think.

**A: We could; however, we think that it is better to keep it in the main text as this information may be important for some readers.**

Figure 6. It’s very difficult to see the uncertainty bars on Figure 6D. could change to black bars?

**A: The color was changed to black.**

L324: Maybe specify that Kolby is “central plains/agricultural”

**A: “An agricultural site” was added (Line 331).**

L350: “Additionally, the chemical composition of the aerosol particles collected by Rosinski et al. (1988) indicate that the air masses in the GoM in July-August were significantly influenced by mineral dust particles.” – wouldn’t presence of dust support Rosinski’s [INP] to be higher than [INP] from this study? Later the authors mention that



the aerosol particles in cold front air masses are likely a mixture of particles from US Central Plains and the GoM – could this be a difference between these measurements and those by Rosinski?

**A: There could be different reasons for the higher [INP] found in the present study in comparison to Rosinski et al. (1988). The presence of mineral dust particles will likely result in a higher [INP]; however, this will also depend on the concentration of dust particles >500 nm, and probably the presence of biological particles. Actually, our recent results (not included in this manuscript) shows that the [INP] in Sisal during the “Saharan dust season” in July 2018 was higher than in winter (Jan-Feb 2017). We believe that differences in the particle size is a key factor. Looking at Figure 4, if we do not take into account the supermicron particles, the total [INP] is lower by one order of magnitude. The following text was added to the revised manuscript (Lines 354-355): “If supermicron particles are excluded, the [INP] at -15 °C from the present study is one order of magnitude lower (Fig. 4)”.**

Figure 7 – suggested change: “influence of the cold fronts (CF)”.

**A: Added.**

L405: what kind of organics? Secondary organic aerosol? Bioparticles are organic.

**A: Secondary organic aerosol was added (Line 418).**