Response to Referee #2

First of all, the authors thank the referee for submitting helpful and meaningful comments, which lead to improvements and clarifications within the manuscript.

Below, we provide our point-by-point responses. For clarity and easy visualization, the Referee's comments (***RC***) are shown from here on in black. The authors' responses (***AR***) are in blue color below each of the referee's statement. In addition to the responses to referees' comments, we further modified the manuscript to increase its clarity and readability. The summary of major and minor changes is included at the end of this document. We introduce the revised materials in green color along/below each one of your response (otherwise directed to the Track Changes version manuscript).

RC: The authors present a set of observation of INP concentrations from rainwater samples collected over West Texas during a 13 months period. They also measure data of precipitation properties, atmospheric temperature, relative humidity and air quality. In addition to that, they performed a metagenomics analysis to obtain information about bacteria present in the rain samples. I personally think that the technical methods used are correctly presented and used. However, the interpretation of their results and the relations they claim be proving between INP and precipitation intensity seems to me completely inaccurate and not backed by their method nor their data at all.

AR: The authors appreciate these general remarks and constructive criticisms regarding our manuscript by Referee #2. We believe that the data presented in the revised manuscript are unique and analysis is robust. We have very good data. The authors sincerely hope the referee considers reading the revised manuscript. We respectfully admit that we have made some insufficient discussions, leading some of our data interpretations in an original manuscript to be speculative. Based on the peer-review comments, we removed/modified them to motivate the research. To allay the reviewer's concerns and mitigate any misgivings, the authors have decided to change the title of manuscript to "**Ice-nucleating particles in precipitation samples from West Texas**", reflecting our changes and articulate what is truly presented in the revised version paper. We have also revised our abstract as well as the conclusion to reflect all of our major revisions (please read the Track Changes version paper). Below, we provide our point-by-point responses in hopes of our manuscript being considered for another review by the reviewer. Please know that problems are not stop signs for the authors. We consider these as important guidelines, and we again thank the reviewer for providing us with ones.

RC: There are 4 major points why I think this paper should be rejected: -Correlation does not imply causality: The intensity of rain is subject to change due to dynamical a thermodynamical factors. For example, I would expect that the large increases in CAPE over the summer season make convective clouds much more intense due to stronger updrafts than those observed on the more stratiform precipitation characteristic of winter like cyclone driven rain. None of these points is addressed minimally in the paper although they are the main drivers of precipitation intensity. INP concentrations can also change seasonally due to a variety of factors (dust transport, dryer conditions, higher biological productivity etc...), such factors are also barely mentioned. Finding a correlation between these 2 variables does not imply any type of causality between them. Also, in case you find a strong correlation, you should attempt to see what direction is this going, is it INP affecting rain or rain affecting INP? You could likely do a similar study with any variable, and you might likely find similar correlations.

AR: We apologize for extending the analysis to interpret the implications towards raised research questions. The authors concur with the reviewer that using our n_{INP} values in precipitation samples collected at the ground level to assess the impact on precipitation properties at cloud height (and vice versa) is not appropriate. Concerning this and many other issues raised by all peer reviewers (e.g., cloud water ≠ precipitation water), the authors decided to substantially revise the manuscript to focus on presenting the observed variation in precipitation properties and somewhat similar $n_{INP}(T)$ in different precipitation systems and their metagenomic analysis. To address what is raised by the reviewer, the authors decided to discuss all caveats (including CAPE) in the new **Sect. 3.5** - Please see the Track Changes version of the manuscript.
"The precipitation intensity strongly depends on several other dynamical factors and thermodynamic conditions, including the land use, moisture levels, land surface temperatures, and convective available potential energy…"

The seasonal variation in aerosol episodes is now discussed as part of **Sect. 3.2**.
"The seasonal variation in PMs may be indicative of different aerosol particle sources or the local meteorological conditions. In the Southern Great Plains, the local sources include harvesting crop fields and agricultural burning..."

The potential impact of dry condition (plus other ambient and precipitation properties) is now discussed in **Sect. 3.1** - e.g.;
"With an overall average of 54.0%, the highest and lowest relative humidity values measured were 70.7 ± 2.3 % (ID# 26; a weak rain sample) and 30.8 ± 0.7 % (ID# 7; a long-lasted rain sample). The observed low ground level relative humidities during some precipitation events (**Tables S1 - S2**) may be a concern as loss of water through partial evaporation of hydrometeors during free fall. But, it is noteworthy that the water evaporation might have negligible effect on n_{INP} estimated from precipitation samples as discussed in **Sect. 2.5**."

The authors revised the text in **Sect. 2.5** to clarify our points regarding CWC as follows.

“We assumed CWC to be a constant of 0.4 g m^{-3}, following Petters and Wright (2015). This assumption would be reasonable for the following three reasons: (1) Petters and Wright (2015) and references therein showed typical values of CWC for different cloud types could narrowly range from 0.2 g m^{-3} to a factor of few more, (2) the authors also showed that the variation of n_{INP} with CWC values for different cloud types in the atmosphere would typically be limited within a factor of two, and our n_{INP} uncertainties could be larger than that, and (3) based on a parametrization for rainwater evaporation, Zhang et al. (2006) suggests that evaporation does not contribute to n_{INP} bias for both strong convective systems and persistent rain events with cloud base heights of ≈3 km. Thus, the variation of CWC on the n_{INP} was considered to be negligible. Nonetheless, it is necessary in the future to further investigate in cloud specific CWCs incorporating with loss of water through partial evaporation of raindrops during free fall based on vertical vapor deficit profiles to conclusively assess if this assumption is fair or not. Precipitation evaporation rate might introduce bias in n_{INP} for precipitation systems with high cloud base, and the correction can be applied accordingly (Petters and Wright, 2015). Direct comparison between INP measurements in cloud water samples and those in precipitation samples might also be key to answer this question (e.g., Pereira et al., 2020).”

Addressing the impact of higher biological productivity on precipitation n_{INP} is a stimulating but tricky task. Agricultural dust is a predominant local dust source in West Texas throughout the year as a number of feedlots exist “within” 33 miles of our precipitation sampling location. Thus, feedlots might act as multiple roles, such as locally emitted INPs, precipitation INPs (if they reach the cloud height), or scavenged aerosol particles. Our result of a dry deposition sample (Sample# 34 – see **Fig. 4**) suggests the limited contribution of local aerosol particles, including feedlot dust. In addition, the authors investigated other dry deposition blank samples (Sample# 35, 38, 39, 40, and 41), and found negligible contribution to $n_{INP.}$ We are not including these results because we ran metagenomics only on Sample# 34. Likewise, our assessment of wet deposition, now presented in the new **SI Sect. S4**, shows negligible impact of scavenged PMs on INPs. As the possibility of feedlot dust entering clouds cannot be ruled out, the authors decided to add the discussion of potential contributions of local agricultural dust to precipitation INPs. We have added the following sentences in **Sect. 3.3**;
“Although we are not certain if these local dusts play a role in precipitation, and assessing the potential of locally emitted aerosol particles to precipitation formation is beyond the scope of the current study, it is important to study the contribution of local agricultural dust in wet scavenging and INP formation at cloud height separately in the future. It is noteworthy that adjacent feedlots (> 45,000 head capacity) are located within 33 miles of our sampling site, and the role of feedlot dusts in atmospheric INPs is described in more detail in Hiranuma et al. (2020). Further discussion regarding the feedlot contribution in INPs in our precipitation samples is provided in **Sect. 3.4**.”

As further investigation in the bioaerosol impact on precipitation n_{INP} is indeed needed, the authors added the following paragraph in our **Sect. 4**;

"We also identified the similarity in bacterial microbiomes between our precipitation and local feedlot dust samples. While we cannot conclude if local feedlot dust contributes to precipitation formation, we find some indications of the inclusion of agricultural dust in our precipitation samples. Regardless, we did not find the previously known bacterial INPs, such as *Pseudomonas* and *Xanthomonas* (Morris et al., 2004) in either the precipitation or feedlot samples. To further seek a connection between local dust and precipitation, it is worthwhile to characterize the local feedlot dust in cloud water samples, as it can be the source of INPs and may impact the local hydrological cycle. Collecting long-term pollen and other biogenic aerosol particles samples and associated observational data for multiple years may add important knowledge regarding the role of local bioaerosols on precipitation INPs."

Please also see our new **Sect. 3.4**;

"…Although we cannot rule out the possibility that scavenging of aerosolized bacteria explains the presence of these bacteria both in feedlot and precipitation samples taken even at a distance from feedlots, our dry deposition background result shows different biological composition (**Fig. 6**). It is also noteworthy to mention that neither of the genera (*Massilia* and *Marinoscillum*) were detected in the background deposition blank sample and it is not known whether they have any IN activity. Therefore, the scavenging may not be the main reason for the presence of *Massilia* and *Marinoscillum* found in our precipitation samples…"

RC: -Wet deposition on rain particles is not properly addressed: Whereas they mention that surface PM does not correlate with INP, this does not discard that wet deposition might be affecting their results.

AR: This is absolutely a valid point. We provide our new interpretation and example to explore potential implications of wet scavenging on our data in **Sect. 3.2** and **SI Sect. S4** and to further motivate the research. Some implications and examples of potential wet scavenging in our INP data are given **in Sect 3.3**. Please see the Track Changes version of the manuscript and SI. Briefly, **Fig. 1** (in the next page) shows the estimated INP concentration of scavenged aerosol particles at four different *T*s, $n_{INP,sv}(T)$, based on scavenged mass simulated with column-integrated mean PM_{10} (see **SI Sect. S4**). We also show the measured INP concentrations of our precipitation samples, $n_{INP,pcpt}(T)$ [L^{-1}], for comparison. As seen in this figure, our estimated $n_{INP,sv}(T)$ values are constantly much lower as compared to $n_{INP,pcpt}(T)$. This trend is true across all ranges of examined *T*s even if we used the ground level PM_{10} as for scavenging inputs. As noted in **Sect. 3.5**, due to many assumptions we made for this analysis, our results of $n_{INP,sv}(T)$ being much smaller than $n_{INP,pcpt}(T)$ may not be conclusive and indeed requires further detailed study. Nevertheless, our estimates suggest the presence of $n_{INP,sv}(T)$ in our precipitation samples. Though the estimated $n_{INP,sv}(T)$ values may be negligible, the authors respectfully take the reviewer's words and removed all discussions associated with influence of INP on precipitation intensity etc.

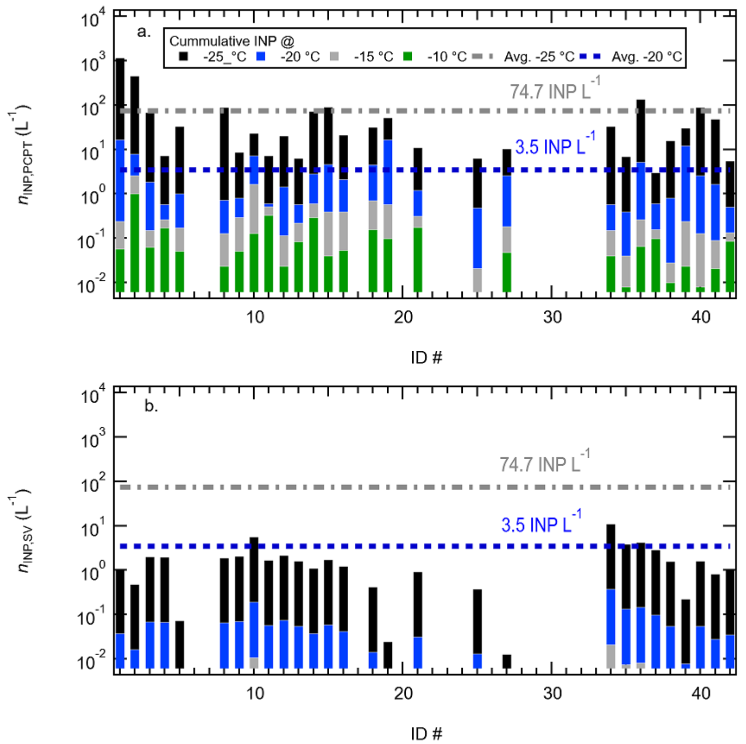

Figure 1. (a) Time series of cumulative n_{INP} (L^{-1} air) in each precipitation sample (ID# shown on the x-axis) at different temperatures. (b) Estimated $n_{INP,SV}$ for a total of 28 samples analyzed based on $M_{sv,cm}$. All data above our n_{INP} detection limit of > 0.006 L^{-1} are shown. The average n_{INP} values at -25 °C (74.7 L^{-1}) and -20 °C (3.5 L^{-1}) in all precipitation samples are shown to guide the reader's eye.

RC: Surface PM is not necessarily a measure of free tropospheric aerosol concentrations, and it is well known that during strong precipitation, aerosol concentrations tend to decrease due to wet deposition. The non correlation between PM and INP is perhaps showing that INP concentrations might be independent on the total aerosol concentration, which is likely given their rareness.

AR: The reviewer is right. We have re-assessed our hourly averaged PM values right before vs. after precipitation (instead of comparing n_{INP} vs. PM measured 'during' precipitation, as previously offered in Fig. S3). Our measurements of PM_1, $PM_{2.5}$, and PM_{10} are summarized in our new **Table 1**. As can be seen in the table, we confirm the trend of PM reduction for all three PM categories after precipitation in part because of scavenging. Therefore, the authors excluded former Fig. S3 and associated discussions from the revised manuscript. The **Sect. 3.2** was revised accordingly. Please see the Track Changes version of the manuscript.

RC: The authors could measure the importance of wet scavenging by analysing the number of particles in their rain samples collected at the surface and just below cloud.

AR: Unfortunately, we do not own cloud water samples. Sampling these as demonstrated in previous studies (e.g., Pereira et al., 2020) and investigating their properties would be an important future work, which is now included in **Sect. 3.5**. Please see the Track Changes version of the manuscript.

An approach of measuring the number of particles in suspension samples to assess the importance of wet scavenging is valid, but requires a hydrodynamic light scattering instrument, which any of the authors do not possess. Instead, as presented above, the authors took a different approach to investigate the "first order" impact of wet deposition - that is, to estimate the amount of scavenged aerosol particle mass, M_{sv} (μg m^{-3}), for each precipitation event using the PM data (i.e., **SI Sect. S4**).

RC: There are strong evidences in their data that point towards wet scavenging being critical, such as how their largest INP concentrations occur on snow samples, which are best at wet scavenging.

AR: This is a valid question. Please see the Track Changes version of the manuscript - **SI Sect. S4**. As discussed in this new section, the estimated scavenging efficiencies of snow are relatively high compared to those of rain as expected (ID# 19 and 21 in **SI Table S6** - almost all PM_{10} scavenged). However, we note that the scavenged PM values of these IDs are not substantially higher compared to those of other rain samples in part due to low measured PM.

RC: -Their statistical analysis is not presented in detail and strongly limited to a few self-selected data samples: The two-sample t-test is a parametric test. Therefore, first they need to show that their distributions are normal, which I think they probably are in logarithmic scale but not on linear scale. Then, they need to present their results clearly and broadly in a reproducible manner, showing the number of data points going in each of the calculations and which dataset are you comparing. Currently they only show the final p-value for a couple of comparisons at high temperature which to me seems not valid at all for a scientific publication. -Their data seems to show many times the opposite to what they claim: Looking at the available data in the supplementary, I can see that intensity of the rain types increases from snow to weak rain to long-lasted rain to hailstorm (being this last one the most intense) (Table S1-3).

AR: We agree. Concerning our limited n_{INP} data (especially thin at -5 °C), we have removed discussions involving p-values and associated with n_{INP}(-5°C). The changes have been reflected in the revised manuscript and SI. Instead, the authors re-analyzed the $n_{INP}(T)$ distribution histogram, categorized based on the season, precipitation type, and precipitation intensity, at -10, -15, -20, and -25 °C. The results are presented in **Figs. 2-4** below. Briefly, we first binned our n_{INP} values at each temperature (i.e., -10, -15, -20, and -25 °C) into five equally sized bins by dividing the n_{INP} range (i.e., max - min) at that temperature by the number six. Subsequently, we visualized the frequency distribution of n_{INP} across different bins on a log scale based on the meteorological season in the U.S. (**Fig. 2**), precipitation type (**Fig. 3**), and maximum precipitation intensity (**Fig. 4**). From these results, we found the followings:

- While no clear seasonal variations of n_{INP} values were apparent in part due to the limited number of samples, the analysis of yearlong ground level precipitation observation as well as INPs in the precipitation samples showed that the highest n_{INP} at -25 °C of 1,130 L^{-1} coincided with a hail-involved severe thunderstorm event in the summer.
- On the other hand, the lowest cumulative INP at the same temperature, 3.2 INP L^{-1}, was found in one of our snow samples collected during the winter.
- Cumulative n_{INP} in our precipitation samples below -20 °C could be high in the samples collected while observing > 10 mm hr^{-1} precipitation with notably large hydrometeor sizes.

These three findings are now included in our main manuscript text. We include these figures in our **SI Sect. S5** to visualize the data in **Tables S1-2**.

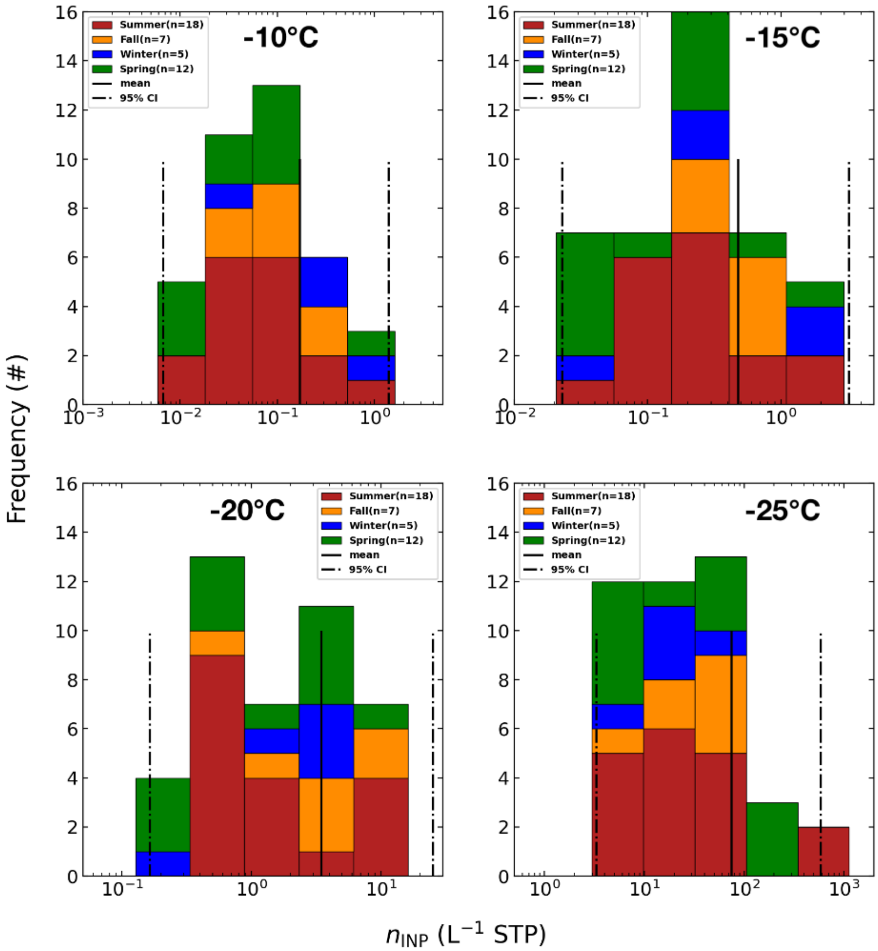

Figure 2. The $n_{INP}(T)$ distribution histogram over different *T*s. The histogram frequency is color-categorized for different meteorological seasons (see **Table S1**). The vertical dashed lines and solid line represent 95% confidence intervals and mean $n_{INP}(T)$ value, respectively.

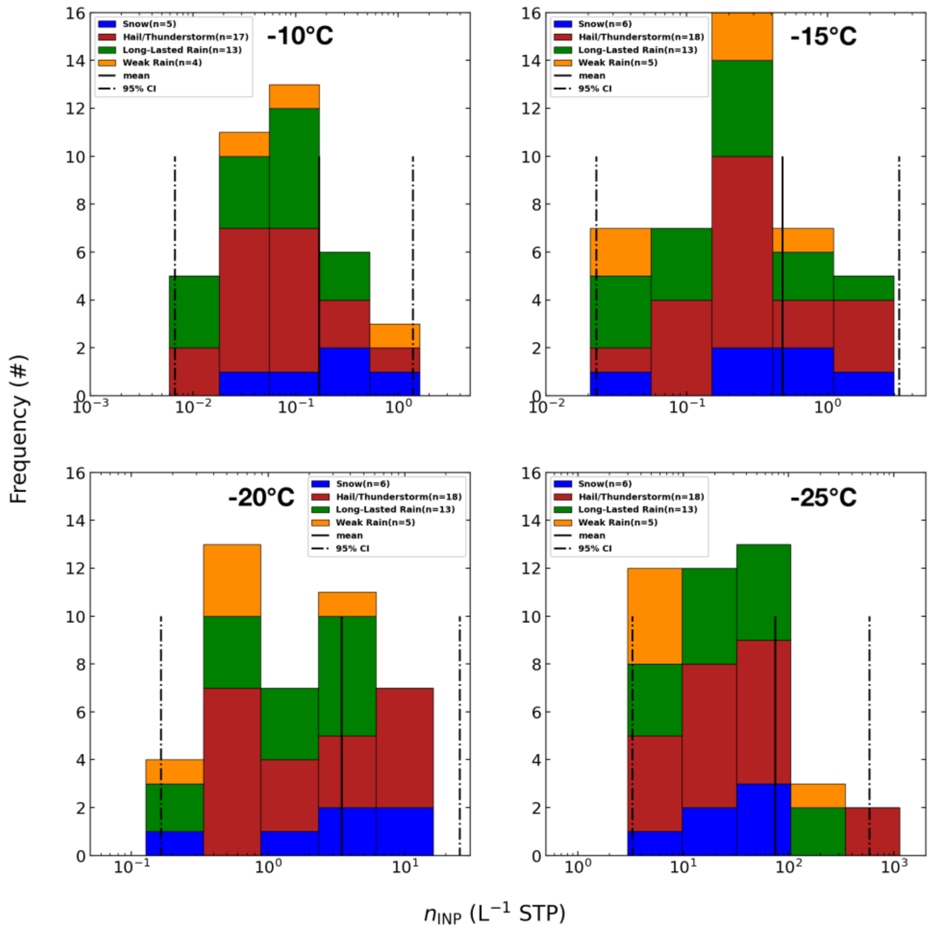

Figure 3. The $n_{INP}(T)$ distribution over different *T*s. The histogram frequency is color-categorized for different types of precipitation, including snow, hail/thunderstorm rain, long-lasted ran, and weak rain, observed at the ground level (see **Table S2**).

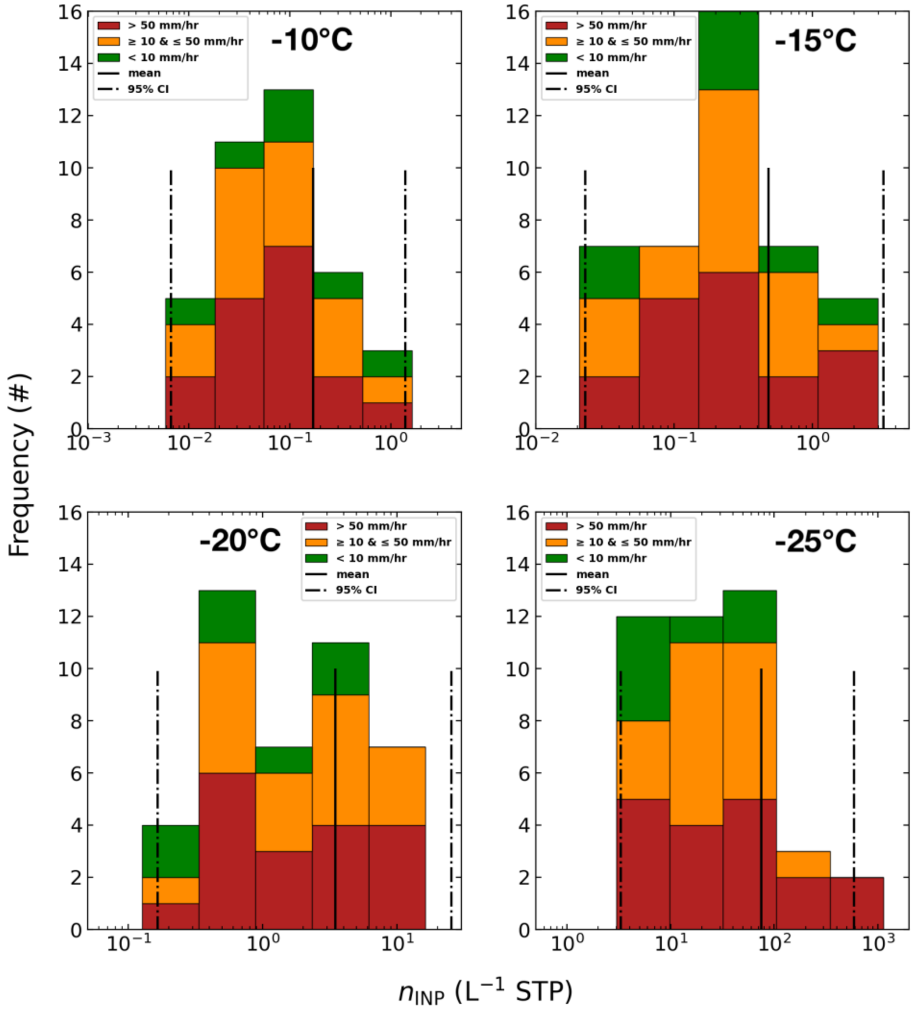

Figure 4. The $n_{INP}(T)$ distribution histogram color-categorized based on three maximum precipitation intensity categories, < 10 mm hr^{-1}, 10-50 mm hr^{-1}, and > 50 mm hr^{-1} (see **Table S2**).

RC: The INP values presented in table S3-1 do not correlate at all with their conclusions, being typically snow the precipitation category with the highest INP measured (at -10, -15, -20 and

-25C) while having the weakest intensity. Of course, this is not the same analysis as performed by the authors, but given the data available in the paper, it seems that the conclusions should, in any case, go the other way around.

AR: The reviewer is right. We removed all insufficient discussion regarding the n_{INP}-intensity relationship. Again, as discussed above, our new interpretation of data only suggests that cumulative n_{INP} in our precipitation samples below -20 °C could be high in the samples collected while observing > 10 mm hr^{-1} precipitation when we observed large hydrometeor size (i.e., Sample# 1 >> Sample# 60 in **Fig. 2b**).

RC: Section 3.2. I like that the authors address the wet deposition factor in this section. However, I do not understand why they relate directly wet deposition with the ambient PM. Wet deposition depends on many factors (size distribution of particles, height from where the droplet falls, etc...)

AR: The authors agree that atmospheric deposition depends on many factors. We now include the wet deposition discussion in **Sect. 3.2** and **SI Sect. S4**. The authors included major caveats and limitations of our study in **Sect. 3.5**. Please see the Track Changes version of the manuscript.

RC: It could have been much more accurate to measure directly the number of particles in each of their precipitation samples.

AR: Measuring the number of particles in our suspension samples to assess the importance of wet scavenging is a valid idea, but requires a hydrodynamic light scattering instrument. Unfortunately, the authors do not own a right instrument, and doing such a rigorous measurements is beyond the scope of the current study.

RC: L366. Whereas a measurable decrease in surface PM during rain suggests a clear removal by wet deposition, a non-measurable decrease in surface PM does not discard wet deposition as the particles could have been absorbed higher up and in amounts below the detection limit.

AR: The authors agree. Our interoperation of wet deposition is presented in **SI Sect. S4**. Please see the Track Changes version of the manuscript.

RC: L393. Snow is a much better scavenger of aerosols than rain. This might be a likely explanation on why you get higher INPs in snow samples. You could test this by measuring the number of particles (and their size distribution) in your snow samples.

AR: Discussed above.

RC: L397-397. I do not see the link here between these 2 ideas.

AR: The authors admit that the analysis/writing was not done properly. We apologize and deleted this sentence.

RC: L426-429. This observation goes against the conclusions of the paper. You observed lower -10C INP values during the May-Aug season when precipitation is stronger (due to the appearance of convective storms) than in the Nov-Jan season.

AR: The authors agree, and this part is removed from the paper.

RC: L430-433 How many points with -5C INPs in the hail/thunderstorm type where included in the analysis. It seems from the plot that there was only 1 point or that all points had the same value. In the supplementary this information is not included.

AR: The reviewer is right about invalidity of these thin data. The discussion on -5 °C INPs is removed in the revised main manuscript.

RC: L440 As per my previous comments, I am not sure how many points are included in this analysis.

AR: Again, the reviewer is right about invalidity of these thin data. The discussion on -5 °C INPs is removed in the revised main manuscript.

RC: L445 what are the results of the statistical analysis over the other temperatures? Showing only the -5C p-values is not enough.

AR: Discussed above.

RC: L447 I don’t think this statement is backed by your results.

AR: Deleted. We apologize for including such an ambiguous statement.

RC: L515-517 Showing some correlations that might be affected by seasonal variations is not enough to claim such a statement.

AR: Deleted. We apologize for including such an ambiguous statement.

Summary of Major Changes

- Our abstract has been revised to reflect all major revisions.
- **Sect. 1.3**: Ambiguous/speculative statements referring to the cloud height condition vs. ground level have been removed; i.e., P3L100-102 and P4L117-120.
- **Sect. 1.4**: Now the study focus is on presenting the ground level observations and measurements, and it is reflected in this particular section with reduced tones.
- **Sect. 3.1**: All repetitive and insufficient statements have been removed or rephrased (e.g., P9L317-322). The authors believe that the readability of this section has improved.
- **Sect. 3.2**: The main focus of this section has been changed to mainly discuss on the wet deposition based on our Air Quality PM sensor data.
- **Sect. 3.3**: Our new data interpretation and comparison to Vali (1968) are now introduced, and our previous statistical analysis has been remove. We re-analyze the n_{INP}, precipitation type observed at the ground level, meteorological season, and precipitation intensity data entirely using histograms (new **SI Sect. S5**).
- **Sect. 3.4**: The authors clarified the connection between feedlot and precipitation samples. We have removed some ambiguous results out of a limited number of samples (i.e., previous Figs. 7b and 7c). All associated texts have been modified, and an unnecessary reference has been removed.
- **Sect. 3.5**: Major caveats and limitations are discussed in this new section. After going through the revision process, the authors realize that including caveats for the reader is as important as offering scientific findings.
- Conclusion is also revised to reflect all major changes addressed above.
- **SI Sect. S4**: Detailed discussion of our interpretation of wet scavenging and its impact on our precipitation INP measurements are discussed in this new SI section. The overview is provided in the main manuscript **Sect. 3.2**.

Minor/technical Changes

- P1 L3: Dimitri → Dimitrios as per request.
- P6 L195-197: The authors realized that removing the frozen fraction ≤ 0.05, accounting for less than 3% of pure water activation (see **Sect. 2.4**), as an artifact shifts our minimum detection to 0.006 L^{-1} for the current study. This detection limit shift has changed a few INP data (but not a substantial amount). The change has been reflected in **Figs. 1-3**, **S1-S2**, and **Table S4**.
- **Sect. 2.4**: Systematic and experimental uncertainties of WT-CRAFT and our experiments are clarified in more intuitive manner.

- **Sect. 2.6**: Identification of our samples for metagenomics is now provided. Note that the precipitation Sample# 50 (another hail/thunderstorm sample) was preserved only for metagenomics.
- **Fig. 2**: Replaced – all the data connecting lines are now removed to increase the visibility of data points.
- Former Fig. 4: Subdivided into two separate figures (**Figs. 4 and 5**) to clarify the associated discussion (new **Fig. 4**: our precipitation INP vs. previous precipitation INP & new **Fig. 5**: precipitation INP vs. local dust INP). All WT-CRAFT data were presented down to -25 °C.
- **Table S1**: Replaced – meteorological seasons are now used to categorize the sampling season instead of previously introduced arbitrary seasonal categories.
- Former Figs. 3, 6, and S3: Deleted as these figures were misleading/oversimplifying the relevant discussion.
- The reference sections have been updated for both main manuscript and SI. The abbreviation sections have been removed as they might not add much value.
- Cory et al. (2019b) and Rodriguez et al. (2020) are removed from the reference list and the main manuscript as Cory et al. (2019a) can represent a single good reference.
- A new reference (Markowicz and Chiliński, 2020) is added for showing an uncertainty of our PM measurements (see **Sect. 2.3**).
- A new acknowledgement is added for useful scientific discussion for the manuscript revision, "We also acknowledge Drs. Gourihar Kulkarni for useful discussions regarding implications of scavenging processes on our data."

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