Response to Referee #1

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: Throughout the course of 13 months the authors have sampled 42 precipitation eventsin the Northwest of Texas and analysed INP concentration in the hydrometeors. Par-allel observations included the size distribution of hydrometeors, airborne particulatematter, air temperature and humidity. Precipitation samples were further subjected tometagenomic analysis, together with a dry deposition sample collected at the samesite and suspended dust samples from a cattle feedlot about 50 km away.

AR: The authors appreciate these general remarks regarding our manuscript by Referee #1. Below, we provide our point-by-point responses. To reflect our changes and articulate what is truly presented in the revised version paper, the authors have decided to change the title of manuscript to "**Ice-nucleating particles in precipitation samples from West Texas**". We have also revised our abstract as well as the conclusion to reflect all of our major revisions (please see the Track Changes version paper).

RC: Data on this variety of parameters was then combined in an interpretation involving numerous implicit and some explicit assumptions, but neglecting two important issues: (a) that surface level air mass on a plain is not necessarily the same as the air mass where precipitation forms (typically "... 2 km to 9 km above ground level ..." (line 66); for vertical gradients in INP concentrations see He et al. (2020)), and (b) that hydrometeors scavenge particles between cloud and ground level. Latter was clearly demonstrated by Hanlon et al. (2017), who produced showers of artificial, sterile rain from a road bridge and collected the artificial hydrometeors, including microbial ice nucleators scavenged during 55 m of free fall, on the field below.

AR: The authors highly appreciate these general and intuitive remarks regarding our manuscript by Referee #1. We also thank the referee for providing us with the references. The reviewer is absolutely right about (a). The INP concentration in general decreases from near ground to cloud height over plains as reported in He et al. (2020). It is clear 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 but somewhat similar $n_{\mathrm{INP}}(T)$ in different precipitation systems and their metagenomics analysis.

The authors agree with the referee's point (b). We provide our new interpretation and example to explore potential implications of wet scavenging on our data in the new **Sect. 3.2** and **SI Sect. S4** and to further motivate the research. Please see the Track Changes version of the manuscript and SI. Briefly, **Fig. 1** below shows the estimated INP concentration of scavenged aerosol particles at four different Ts, $n_{\mathrm{INP,sv}}(T)$, 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, $n_{\mathrm{INP,pcpt}}(T)$ [L^{-1}], for comparison. As seen in this figure, our estimated $n_{\mathrm{INP,sv}}(T)$ values are constantly much lower as compared to $n_{\mathrm{INP,pcpt}}(T)$. This trend is true across all ranges of examined Ts 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 $n_{\mathrm{INP,sv}}(T)$ being much smaller than $n_{\mathrm{INP,pcpt}}(T)$ may not be conclusive and indeed requires further detailed study. Nevertheless, our estimates suggest the

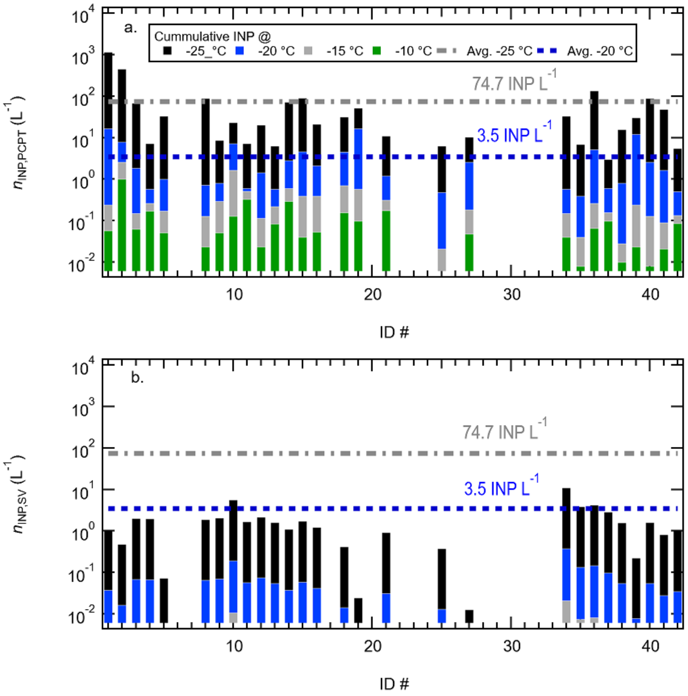

Figure 1. (a) Time series of cumulative n_{INP} (L^{-1} air) in each precipitation sample (ID# shown on the x-axis) at different temperatures. (b) Estimated $n_{\mathrm{INP,SV}}$ for a total of 28 samples analyzed based on $M_{\mathrm{sv},cm}$. All data above our n_{INP} detection limit of > 0.006 L^{-1} 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.

presence of $n_{\mathrm{INP,sv}}(T)$ in our precipitation samples. Though the estimated $n_{\mathrm{INP,sv}}(T)$ 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.

RC: Figure 4 exemplifies the problem I see with the combination of little-related data and ignored processes. [1] The Figure combines INP data on airborne dust samples near ground (feedlot, 50 km away from other observations), [2] INP estimates of atmospheric INP concentrations at cloud height derived from precipitation samples and an assumed cloud water content (ignoring scavenging of particles and loss of water through partial evaporation of raindrops during free fall (ground level RH during rainfall 31% to 71% (line 309)), and [3] an atmospheric INP estimate based on a dry deposition sample suspended in an (arbitrary?) volume of pure water and transformed into an atmospheric concentration value. I think the data from these three kinds of sources cannot be directly compared because of mentioned issues.

AR: RE [1]: 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** in the revised manuscript) suggests the limited contribution of local aerosol particles, including feedlot dust. Likewise, our assessment of wet deposition, now presented in **SI Sect. S4**, shows $n_{\mathrm{INP,PCPT}}(T)$ being much larger than $n_{\mathrm{INP,sv}}(T)$. As the possibility of feedlot dust entering clouds cannot be ruled out, the authors would like to retain the discussion of potential contributions of local agricultural dust to precipitation INPs. To clarify our point of including feedlot dust INP data, 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**."

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…"

AR: RE [2]: The authors revised the text in **Sect. 2.5** to clarify our points regarding CWC as follows.

"We presumed CWC to be a constant of 0.4 g m^{-3}, covering the continental clouds in our study. Our assumption would be reasonable since Petters and Wright (2015) 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. Thus, the effect of CWC on the n_{INP} would be negligible."

→

"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)."

AR: RE [3]: Thanks for asking. The volume of pure water used to assess our dry deposition sample (Sample# 34), which is 5 mL, was arbitrarily determined by averaging collected precipitation volumes of all prior samples (Sample# 1 to 33). A new sentence is now added in P4L141 to clarify this as follows:

"We note that a volume of pure water (5 ml) for an atmospheric INP estimate based on a dry deposition sample was determined by averaging collected precipitation volumes of all samples prior to this dry deposition sample."

RC: However, the paper definitively contains new and interesting observations that may be interpreted to a certain extent, without making too many implicit or explicit assumptions. These observations are foremost the INP concentrations in precipitation samples combined with the precipitation properties, including kind of precipitation, size spectra of the hydometeors, precipitation duration and intensity. Such an interpretation needs to address the issues of below-cloud scavenging and also the higher scavenging efficiency of snow as compared to rain (Wang et al., 2014).

AR: Thank you – We also believe that the data presented in the revised version manuscript are unique and analysis is robust. We have very good data. We have improved the clarity of precipitation properties and how we interpret potential impacts of scavenging on our data etc. in **Sects. 3.1, 3.2, and S4** (please see the Track Changes version paper and SI). As discussed in these sections, 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 scavenged). However, we note that the M_{sv} values of these IDs are not substantially higher compared to those of other rain samples in part due to low M_0. Some implications and examples of potential wet scavenging in our INP data are given **in Sect 3.3**.

Our finding on maritime bacteria in West Texas adds an important caveat for the precipitation INP study – a link between microphysics and dynamics beyond regional scale. The authors now extended this discussion in **Sect. 3.4** (please see the Track Changes version paper). The authors indeed wish to continue including this part in the manuscript. We now included the discussion of local and long-range PM sources/transport in **Sect. 3.2**.

RC: In contrast, data on particulate airborne matter near ground level is something I would put aside when revising the manuscript.

AR: The authors agree and deleted former Fig. 3. We also excluded the PM data collected during precipitation from **SI Table S2**. We note that we used our PM data collected before precipitation to assess the scavenging efficiency of PMs and its impact/implication on our precipitation INP estimation.

RC: I found it tedious to read through listings of data in the Results and Discussion section. Somehow, I missed a clear storyline. It would have been a more engaging reading experience, if Figures were not introduced by full sentences that resemble Figure legends.

AR: The authors apologize for all of our confusing and cumbersome statements, resulted in an unclear story, in the original discussion manuscript. We gave careful re-interpretation of our data and revisions to remove all logical leaps and insufficient discussions.

RC: To give an example (lines 372 and following): "Figure 4 shows the IN spectra for different precipitation types analyzed in this study superposed on the IN spectral boundaries adapted from a previous precipitation INP study (Petters and Wright, 2015). This figure also displays other reference IN spectra, including our 24-hour dry deposition blank sample (collected from January 2 – 3, 2019 at our sampling site) and IN spectra measured for dust suspension samples collected from the downwind side of a local feedlot (identity purposely concealed), where substantial and consistent dust emission historically persists (Whiteside et al., 2018). For the measured T range, nINP values from dry deposition blank sample were at least an order of magnitude lower than that from our precipitation samples." This entire section could simply be replaced with: "For the measured T range, nINP values from dry deposition blank sample were at least an order of magnitude lower than that from our precipitation samples (Figure 4)."

AR: The authors took the reviewer's word for it. Thank you.

RC: What is meant by (line 313): "...substantial number of precipitation particles with a cumulative number of 2E+05 to 6.6E+05 per event." Perhaps "...precipitation particles recorded by the disdrometer...", or "...precipitation particles per square metre..."?

AR: The former of the reviewer's comments is correct. These are the absolute number of precipitation particles passing through the laser beam cross section of and detected by our disdrometer. We have rephrased the manuscript text as follows;
"In our study period, a disdrometer detected a substantial number of precipitation particles with a cumulative number ranging from 1.0×10^4 to 6.6×10^5 particles passing through its laser beam cross section per event."

RC: Lines 323-327: It is not clear why the range of intensities is indicated as "0 to 150 mm hr-1", when maximum intensity was 129.3 mm hr-1 and minimum intensity 1.1 mm hr-1?

AR: Thank you for catching this. The numbers are corrected.

RC: References: Whiteside et al. 2018: I would have liked to learn more about this study, but could not find it. A link to the paper, if available, would have been useful.

AR: As per request, the authors provide the doi link of Whiteside et al. poster.

> Whiteside, C. L., Auvermann, B. W., Bush, J., Goodwin, C., McFarlin, R., and Hiranuma, N.: Ice nucleation activity of dust particles emitted from cattle feeding operations in

> the Texas Panhandle, Poster, AMS - 10th Symposium on Aerosol-Cloud-Climate Interactions, Austin, TX, USA, doi: 10.13140/RG.2.2.29505.38248, 2018.

The authors, however, note that more exclusive feedlot INP data (over 2016-2019) generated using the same immersion freezing assay has recently become available in the following ACPD (e.g., Fig. 3):

> Hiranuma, N., Auvermann, B. W., Belosi, F., Bush, J., Cory, K. M., Fösig, R., Georgakopoulos, D., Höhler, K., Hou, Y., Saathoff, H., Santachiara, G., Shen, X., Steinke, I., Umo, N., Vepuri, H. S. K., Vogel, F., and Möhler, O.: Feedlot is a unique and constant source of atmospheric ice-nucleating particles, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1042, in review, 2020.

Since the data presented in Whiteside et al. 2018 are adapted and merged in this new manuscript, we have replaced Whiteside et al. with Hiranuma et al.

RC: Gabor Vali determined INP spectra in rain and hail samples from numerous storms in various parts of North America (Vali, 1968). The authors may find it helpful to have a look at his work when revising their manuscript.

AR: The authors appreciate the referee for providing a useful reference. It indeed helped us in better understanding the previous INP measurements from hail and rain samples. We have modified our **Fig. 4**, and added the following discussions in the new **Sect. 3.3**;
"**Figure 4** shows a compilation of $n_{INP}(T)$ spectra of each precipitation type in comparison to previously reported precipitation $n_{INP}(T)$. In general, most of n_{INP} spectra fall in the upper range of the previous precipitation n_{INP} data presented in Petters and Wright (2015) and Vali (1968). INP humps shaping the reference spectra (i.e., one below -20 °C and another at > -20 °C) are also found in our spectra. The observed hump is especially obvious for n_{INP} at T above -20 °C, and some of our spectra exceed the upper bound of the reference spectra in any precipitation types. For Ts below -20 °C, our $n_{INP}(T)$ data match fairly well within the range of the reference $n_{INP}(T)$ for all four precipitation types. Thus, the precipitation type observed at the ground level would not have any relationships with INP propensity at least for our 42 samples collected for this study. However, it is interesting that most of our n_{INP} data points above -15 °C fall within the range of estimated n_{INP} at cloud height with < 50% storm efficiency, reported in Vali (1968). In fact, regardless of precipitation type, we see reasonable overlaps of our $n_{INP}(T)$ with Vali (1968). The author stated that the large differences in IN content among precipitation samples were mainly caused by differences in the nucleus content of the air entering the storm. This implies that the cloud level dynamics like cloud entrainment impact the cloud level INP concentrations. Hence, we compared our precipitation INP data with the

lower and upper limits of the IN concentrations in the air entering the storm given by Vali (1968) (Table 2, Chapter# 9). These cloud level INP concentrations given by Vali (1968) were for two different storm efficiencies, which is the ratio of mass of precipitation to the mass of water input. The storm efficiency of 10% represents the time when high concentrations of precipitation inside the storm begins to develop. Likewise, 50% is at the peak intensity of the storm. These different combinations of storm efficiencies and water content accounted for a tenfold variation in the ice nucleus content. As more air is entered into the storm with 50% efficiency, more IN concentrations are observed at cloud level. Though our data are comparable to Vali (1968), there is still indeed the need for cloud level INP measurements to define the relationship between the ground level INP concentrations and precipitation intensity."

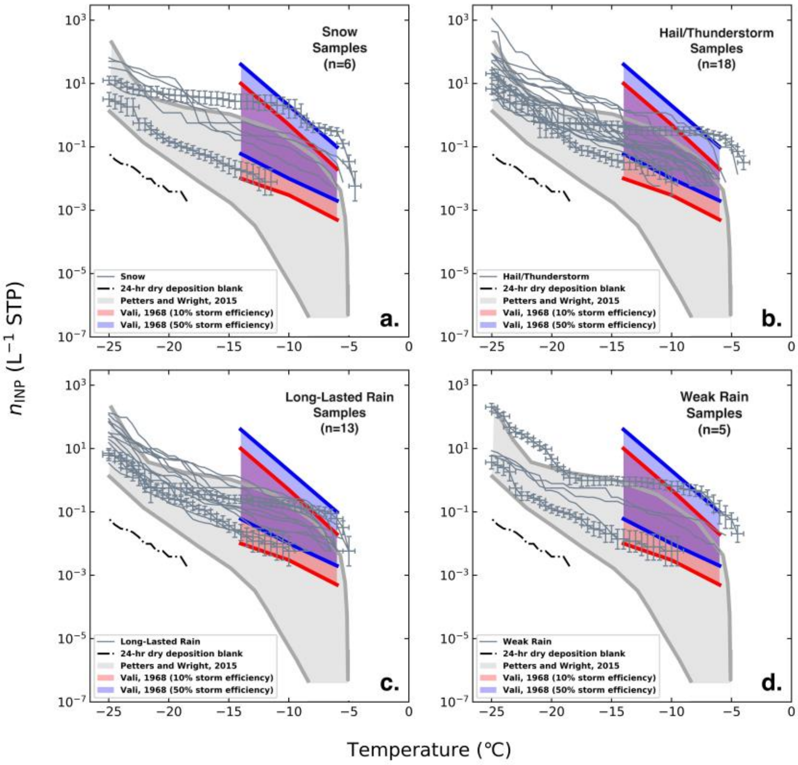

Figure 4. IN spectra of (a) Snow, (b), Hail/Thunderstorm, (c) Long-Lasted rain, and (d) Weak rain samples superposed on nucleation spectra from previous precipitation INP studies (shaded areas). A subset of spectra shows error bars. The X-axis error bars represent constant uncertainty of ±0.5 °C in temperature. The Y-axis error

bars are the 95% confidence interval for n_{INP} shown only for two samples from each category. The number of precipitation samples in each category is shown by the value of 'n'.

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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