Response to Referee #3

The authors would like to express our sincere gratitude for the referee and helpful comments. Below, we provide our point-by-point responses. The referee's comments (RC) are shown from here on in black. The authors' responses (AR) are in blue below each of the referee's statements. We introduce the revised materials in green color along/below each one of your responses (otherwise directed to the Track Changes version manuscript). All references are available at the end of this AR document.

General comment:

RC: This is a valuable contribution to the ice nucleation community and it has a good level of novelty. This is the first time that I reviewed this manuscript and given that this was the second round of revisions I was expecting to see a more readable, clean, and concise document. Although the incorporated changes improved the manuscript, it cannot be accepted in its current version. I invite the authors to take into account the comment by original reviewer #2 "I do not wish to spend a great deal of time going from line to line trying to edit the manuscript for them".
AR: The authors appreciate these general remarks. We reconsidered previous referee #2 comments and revised our manuscript, especially the Materials and Methods section, accordingly. Below, the authors provide our point-by-point responses.

Major Comments:

RC: The authors wanted to cover too much: i) field (annual trends, seasonal trends, downwind vs, upwind); ii) field vs. lab; iii) lab (heat vs. no heat, super micron vs. sub micron, bulk vs. filter,); iv) intercomparison between 4 different instruments; and v) a parametrization. Although it is not bad at all to be ambitious and to report several measurements/observations, the authors need to combine the large amount of data into something that is easy to read and to follow.
AR: The authors agree with the referee. We realized that the structure of the method section and presenting various things could be overwhelming. The authors also acknowledge that retaining the format from a previous version would cause additional confusion. Therefore, the authors decided to restructure the main article to present the most invaluable scientific outcomes and limit the amount of Supplemental Information (SI) for the sake of readability. Our decision is based on considering all four referees' comments.

The authors revised the manuscript to feature the **abundance of supermicron aerosol particles acting as feedlot ice-nucleating particles (INPs) from lab and field studies** and did the following modifications:

(1) Separating **Sects. 2 and 3** based on laboratory study (sub-section 1) and field investigation (sub-section 2) to explain methods, materials, and results for each sub-section independently.
(2) Moving sample descriptions to the Results and Discussion section (**Sect. 3.1.1**) and leaving only concise technique explanation in the Materials and Methods section (**Sect. 2.1.1**) to increase the readability of the manuscript in an organized manner.
(3) Moving the heat treatment data, outcomes, and discussion from the main manuscript into a single **SI Sect. S4**. Keeping it over different sections in methods and results impaired the overall readability in our previous version, and the authors believe that this modification resolves the readability issue.
(4) Removing all bulk sample discussions from this manuscript and focusing on the filter-collected aerosolized samples – So there will be no bulk vs. aerosolized sample discussion. In the end, the

bulk is not our main outcome. The revised paper focuses on lab vs. field, we believe that the aerosolized sample is more relevant to what is in the field than the bulk sample.

(5) Removing the ice residual composition discussion from the main manuscript. We do not have statistically valid aerosol particle composition data of our 'field' samples from this study. As the referee is concerned, we cannot conduct the comparison of laboratory and filed sample compositions, and the former Sect. 3.4 contributes little to the main text. The authors agree that the composition analysis is not the main focus of this manuscript, and decided to exclude this part from the manuscript.

(6) Moving the discussion regarding the estimated INP concentrations to **SI Sect. S5**.

In addition, the authors also changed the title of our manuscript to "Laboratory and field studies of ice-nucleating particles from open-lot livestock facilities in Texas", which better represents our research.

RC: Given that the samples were found to be heat insensitive, the IN properties are likely coming from the mineral components. Why the mineralogical composition is not reported. In lines 110-112 it is stated "However, our knowledge regarding what particular features of OLLF dust trigger immersion freezing at heterogeneous freezing temperatures (Ts; i.e., size vs. composition) is still lacking". The authors focused on the size of the particles but in terms of composition the minerals are completely ignored.

AR: The authors conclude that investigating minerals is not relevant to understand feedlot-derived INPs in this study because of the following reasons:

- Our previous work using Raman micro-spectroscopy revealed that ≈ 96% of ambient aerosol particles sampled at the downwind edge of an open-lot livestock facility (OLLF) contain brown or black carbon, hydrophobic humic acid, water-soluble organics, less soluble fatty acids, and carbonaceous materials mixed with salts and minerals (Hiranuma et al., 2011). We have mineralogical understanding, but we miss the bulk composition information (e.g., X-ray fluorescence and X-ray diffraction).
- Our chemical composition analysis of laboratory samples (**SI Sect. S1**) indicates that our samples are exclusively organic in nature in terms of aerosol composition.
- Recently, organic acids (i.e., long-chain fatty acids) and heat-stable organics were found to act as efficient INPs (DeMott et al., 2018; Perkins et al., 2020). Thus, identifying heat-stable organic compounds and studying their physicochemical properties may be key to understand the properties of OLLF INPs.
- A comparison of our field ice nucleation active-surface site density ($n_{s,geo}$) data to ice nucleation (IN)-active minerals (e.g., K-feldspar, quartz) does not support the inclusion of IN-active minerals. Some of our field samples (from any season) contain more efficient INPs than K-feldspar at temperatures above approximately -15 °C, as seen in Figure 1 on the next page. The $n_{s,geo}$ spectra of our field data generally have gentler slopes than K-feldspar. We note that the immersion freezing efficiency of K-feldspar is higher than quartz (Atkinson et al., 2013; Harrison et al., 2019).

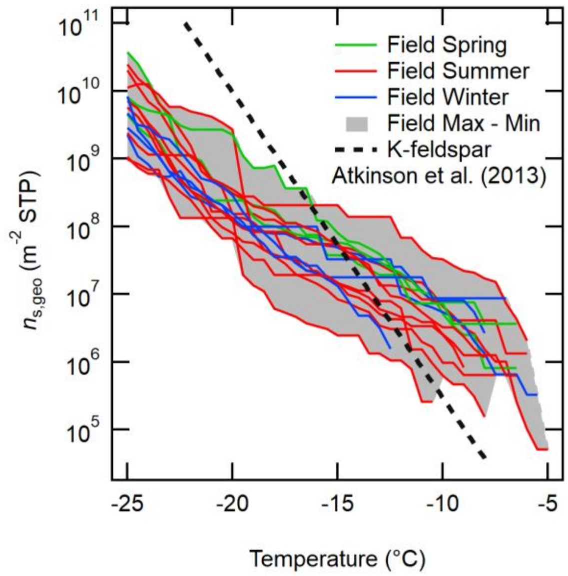

Figure 1. Comparison of our field $n_{s,geo}$ data (adapted from Fig. 8 in our revised manuscript) to K-feldspar. The $n_{s,geo}$ values of K-feldspar were derived using the laser diffraction-based surface-to-mass ratio, 0.89 m^{-2} g^{-1}, and Brunauer-Emmett-Teller (BET) specific surface area, 3.2 m^{-2} g^{-1}, reported in Atkinson et al. (2013).

In addition, we could not find any notable inclusions of known IN-active microbiomes in both laboratory and field samples (now discussed in **Sect. 3.3.1**). While we cannot rule out the possibility of IN from TXD01 and TXD05 samples triggered by biological INPs, our current results do not support it. Certainly, our study cannot conclude what particular features of OLLF dust trigger immersion freezing at heterogeneous freezing temperatures. However, this deficit is a good motivation to investigate OLLF-derived ice crystal residual samples in more detail in the future.

RC: This is the second round of revisions and the manuscript still needs to be edited to improve its readability and the language. I was expecting to see a cleaner version.
AR: The language is re-checked by an English native speaker and an external editorial service provider. Please see the track changed materials. With a revised paper structure, the authors believe that the readability is improved.

RC: Lines 367-368 it is mentioned that "These results imply the following: (1) ambient meteorological conditions, as summarized in Table 1, might not be determining factors for nINP for our study sites". This refers to RH, T and P but not to wind speed, one of the most important meteorological variables to resuspend dust particles. Why is wind speed and wind direction not reported?
AR: Wind direction was not reported because we arranged our sampling locations (i.e., downwind and upwind sites) according to the observed wind direction. For instance, when south wind prevailed (90° < wind direction < 270°), we used the Northern site as the downwind site. Likewise, the Southern site was used as the downwind site while the north wind was dominant (270° < wind direction < 90°). We include this point in the revised **Sect. 2.2.1**. The authors provide our observed wind properties, which include wind speed and direction, during our sampling activities in Table 1 on the next page.

Table 1. Average wind speed and direction (± standard deviation) for individual sampling activities.

Year	Date	Location	Start Time (Local)	End Time (Local)	Average Wind Speed ± Standard Deviation (mile hr^{-1})	Average Wind Direction ± Standard Deviation (degree)
2019	20190715	OLLF-1	18:45:00	22:05:00	3.6 ± 1.3	157.9 ± 13.9
	20190716	OLLF-2	18:45:00	20:29:00	10.6 ± 1.7	186.4 ± 4.3
	20190724	OLLF-3	19:24:00	20:34:00	10.1 ± 1.3	147.5 ± 6.6
	20190226	OLLF-1	16:08:00	19:09:00	11.2 ± 4.3	207.9 ± 13.2
	20190328	OLLF-2	16:26:00	20:52:00	8.7 ± 3.3	217.2 ± 6.7
	20190420	OLLF-3	17:05:00	21:05:00	10.2 ± 2.9	197.2 ± 19.1
	20190116	OLLF-1	16:03:00	19:33:00	16.6 ± 2.8	256.0 ± 6.8
	20190117	OLLF-2	15:48:00	19:30:00	8.7 ± 1.8	188.3 ± 11.6
	20190118	OLLF-3	15:40:00	18:40:00	23.3 ± 2.5	319.4 ± 33.1
2018	20180722	OLLF-1	18:42:00	22:39:00	5.7 ± 1.6	170.7 ± 11.0
	20180723	OLLF-2	18:42:00	22:17:00	5.1 ± 3.9	83.6 ± 21.1
	20180724	OLLF-3	18:20:00	22:13:00	7.9 ± 1.9	136.6 ± 12.0
	20180416	OLLF-4	4:53:30	8:06:40	12.1 ± 4.0	216.2 ± 8.3
2017	20170709	OLLF-1	19:32:45	22:26:00	9.3 ± 2.9	160.5 ± 10.1
	20170710	OLLF-2	18:06:00	22:06:30	10.3 ± 3.0	183.8 ± 9.0
	20170711	OLLF-3	18:28:00	22:08:00	6.4 ± 1.7	172.0 ± 10.9

Resuspension of feedlot surface materials, the so-called hoof action, is not wind-driven. Cattle movement and hoof action are the decisive emissions mechanism of feedlot dust when the air is dry and hot as described in Auvermann (2001) and Bush et al. (2014). The authors performed linear regression analysis for wind speed vs. particulate matter (PM) concentration, and the resulting Pearson correlation coefficient (*r*) was -0.32 (see Table 2 on the next page). Concerning highly variable concentration, the authors also examined the relationship between wind speed and cumulative PM mass per unit time, and the resulting *r* was -0.35. Since these negative coefficients indicate an inverse relationship between wind speed and PM, the authors decided not to report it. Nevertheless, we now clarify this point in the revised **Sect. 3.2.2** and report both wind speed and direction data activities in **Table 7**.

Table 2. Summary of wind speed, average PM mass concentration, and cumulative PM mass for individual sampling activities.

Year	Date	Location	Start Time (Local)	End Time (Local)	Wind Speed (mile hr^{-1})	Average PM Mass Concentration (mg m^{-3})	Cumulative PM Mass (µg hr^{-1})
2019	20190715	OLLF-1	18:45:00	22:05:00	3.63	0.21	50.46
	20190716	OLLF-2	18:45:00	20:29:00	10.58	0.09	24.17
	20190724	OLLF-3	19:24:00	20:34:00	10.07	0.33	90.00
	20190226	OLLF-1	16:08:00	19:09:00	11.24	0.13	18.96
	20190328	OLLF-2	16:26:00	20:52:00	8.66	0.15	46.13
	20190420	OLLF-3	17:05:00	21:05:00	10.22	0.05	8.63
	20190116	OLLF-1	16:03:00	19:33:00	16.63	0.01	3.43
	20190117	OLLF-2	15:48:00	19:30:00	8.74	0.05	11.22
	20190118	OLLF-3	15:40:00	18:40:00	23.30	0.35	83.93
2018	20180722	OLLF-1	18:42:00	22:39:00	5.71	0.82	324.30
	20180723	OLLF-2	18:42:00	22:17:00	5.12	2.48	814.30
	20180724	OLLF-3	18:20:00	22:13:00	7.88	0.22	86.03
	20180416	OLLF-4	4:53:30	8:06:40	12.13	0.03	12.08
2017	20170709	OLLF-1	19:32:45	22:26:00	9.26	0.49	154.22
	20170710	OLLF-2	18:06:00	22:06:30	10.31	0.18	56.51
	20170711	OLLF-3	18:28:00	22:08:00	6.42	0.15	46.77

RC: Lines 417-418 and Line 524: "For each sample, the spectra nearly overlap each other at T ~ -25 °C, verifying their comparability and complementing features." and "Upon confirmation of the comparability between field and lab ns,geo values". Why is this overlap only true at -25C and not at other temperatures? obtaining similar results at this temperature is enough to conclude that the systems are really comparable. Is it not the overlap at -25C suspicious? Why should they be comparable? In the field, aerosol particles are naturally aerosolized and this resuspension from the ground can favor specific aerosol sizes. On the other hand, the aerosolization in the laboratory is mechanical and the particles sizes, and hence, their composition may significantly differ from those found in natural environments.

AR: The authors understand the referee's concern. The authors intended to point out (1) our dynamic filter processing chamber (DFPC)-derived $n_{s,geo}$ values in **Fig. 4** agreed reasonably well with the ice nucleation spectrometer of the Karlsruhe Institute of Technology (INSEKT) results at the measured temperatures within our error ranges, and (2) both aerosol interaction and dynamics in the atmosphere (AIDA) and INSEKT show reasonably comparable results in the overlapping temperatures around -25 °C. These two ambiguous statements are now removed, and the potential source of discrepancy between laboratory and field results is now discussed in the revised **Sect. 3.3.1**.

RC: Lines 438-441: The authors need to show that the composition and particle size distribution of ambient and Laboratory particles are comparable.

AR: Upon the request, the authors conducted energy dispersive X-ray spectroscopy (EDX) on particles in the suspended field sample, collected from OLLF-1 on July 22, 2018. We used an electron microscope (JEOL, JSM-6010LA) equipped with an EDX function. We have looked at a total of 56 particles on an aluminum substrate. All particles had an area equivalent diameter smaller than 6.44 µm, which is the largest aerosol particle size found in the AIDA chamber (see **Table 1**). Then, we qualitatively assessed EDX signals of organic (C, N, O), salt-rich (Na, Mg, K, P), mineral-rich (Si, Ca), and others. We excluded a background signal of aluminum from a substrate. We detected carbon in all particles exclusively with the inclusion of minerals for 20% of examined particles. Two representative electron microscopy images and EDX spectra are shown below in Fig. 2.

The observed predominance of carbonaceous particles is consistent with our previous field sample analysis from the same OLLF (Hiranuma et al., 2011). We also echo that our single particle mass spectrometry composition analysis of laboratory samples (**SI Sect. S1**) indicates that our samples are exclusively organic in terms of aerosol composition.

Note that, as stated in the Materials and Methods section, our field sample was consumed for immersion freezing analysis within a day after sampling concerning sensitivity to storage time. Thus, our composition result may not reflect the composition analyzed for the West Texas cryogenic refrigerator applied to freezing test system (WT-CRAFT) for immersion freezing. Moreover, our EDX analysis on a small number of particles from a single sample cannot provide any statistically valid conclusion and size distribution data. Concerning these deficits and the referee's comment, the authors decided to omit the discussion of particle composition from the main manuscript.

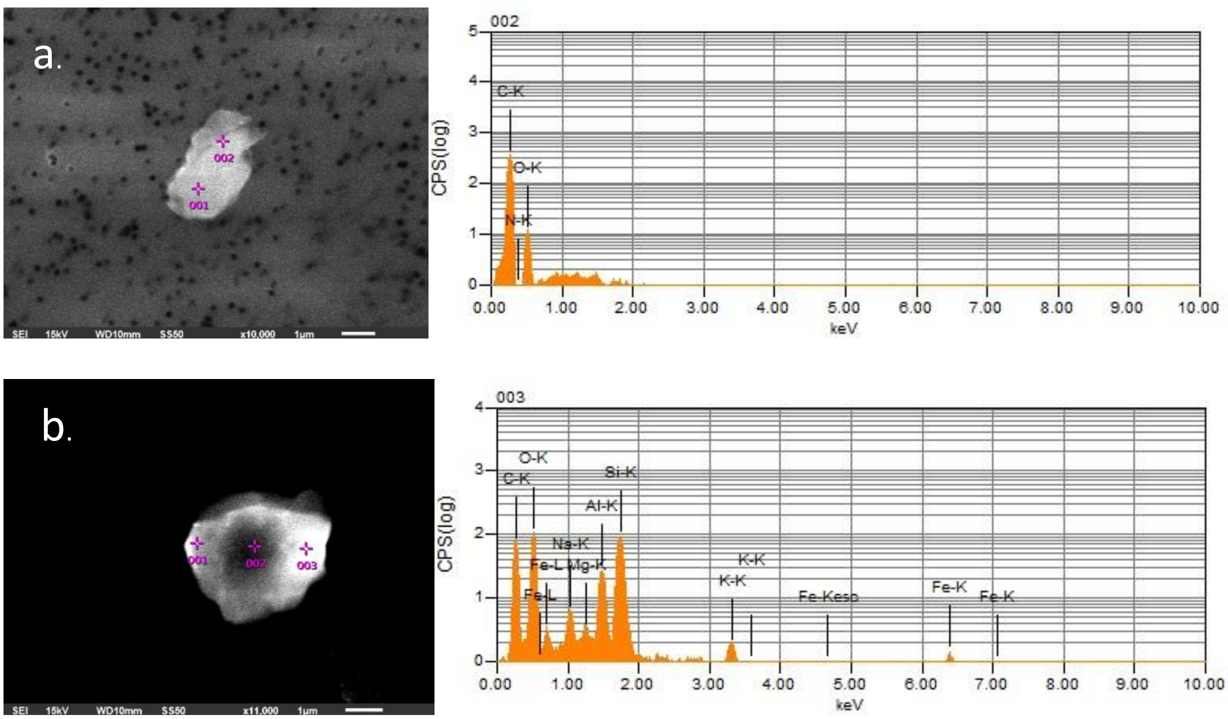

Figure 2. Typical electron microscopy images and EDX spectra of (a) organic dominant particle (ID# 12) and (b) mineral including particle found in the OLLF-1 sample (ID #1).

As demonstrated in our previous study, the surface area distribution of ambient OLLF dust peaks in mode diameter at ≤ 10 µm (Hiranuma et al., 2011). This mode diameter is larger than surface-derived samples aerosolized and examined in the AIDA chamber. However, it is cautiously noted that the ambient OLLF dust size distribution is not spatially uniform, and the emitting mechanism itself is not controllable as it highly depends on a unit of mobile livestock. Granting the primacy of hoof action as the decisive emissions mechanism of OLLF dust as described in Bush et al. (2014), a more controlled laboratory experiment has been desired to characterize IN ability of OLLF soil dust.

Minor Comments:

RC: The Abstract is extremely long. The authors need to shorten it focusing on the main results only. Similar to the Abstract, several parts in the Results section (and along the manuscript) are repetitive, longer than needed and not very concrete. The manuscript needs to be further edited to improve its readability.

AR: The abstract is revised, and the text count was reduced from 601 words (3952 characters) to 384 words (2600 characters). All repetitive parts are removed from the manuscript. The revised manuscript focus on the **abundance of supermicron aerosol particles acting as feedlot INPs from lab and field studies** in a concise and readable manner. Below is the revised abstract.

"In this work, an abundance of ice-nucleating particles (INPs) from livestock facilities was studied through laboratory measurements from cloud simulation chamber experiments and field investigation in the Texas Panhandle. Surface materials from two livestock facilities, one in the Texas Panhandle and another from McGregor, Texas, were selected as dust proxies for laboratory analyses. These two samples possessed different chemical and biological properties. A combination of aerosol interaction and dynamics in the atmosphere (AIDA) measurements and offline ice spectrometry was used to assess the immersion freezing mode ice nucleation ability and efficiency of these proxy samples at temperatures above -29 °C. A dynamic filter processing chamber was also used to complement the freezing efficiencies of submicron and supermicron particles collected from the AIDA chamber. For the field survey, periodic ambient particle sampling took place at four commercial livestock facilities from July 2017 to July 2019. INP concentrations of collected particles were measured using an offline freezing test system, and the data were acquired for temperatures between -5 °C and -25 °C.

Our AIDA laboratory results showed that the freezing spectra of two livestock dust proxies exhibited higher freezing efficiency than previously studied soil dust samples at temperatures below -25 °C. Despite their differences in composition, the freezing efficiencies of both proxy livestock dust samples were comparable to each other. Our dynamic filter processing chamber results showed on average approximately 50% supermicron size dominance in the INPs of both dust proxies. Thus, our laboratory findings suggest the importance of particle size in immersion freezing for these samples, and that the size might be a more important factor for immersion freezing of livestock dust than the composition. From a three-year field survey, we measured a high concentration of ambient INPs of 1,171.6 ± 691.6 L^{-1} (average ± standard error) at -25 °C for aerosol particles collected at the downwind edges of livestock facilities. An obvious seasonal variation in INP concentration, peaking in summer, was observed with the maximum at the same temperature exceeding 10,000 L^{-1} on July 23, 2018. The observed high INP concentrations suggest that a livestock facility is a substantial source of INPs. The INP concentration values from our field survey showed a strong correlation with measured particulate matter mass concentration, which supports the importance of size in ice nucleation of particles from livestock facilities."

RC: Avoid self citations. From my personal point of view this is excessive.

AR: By limiting our research focus as addressed above, self-citation has been reduced. However, the authors keep all essential, meaningful citations as-is.

RC: Move sections 3.3 and 3.4 to SI as they contribute little to the main text.

AR: The authors moved some technical details of metagenomics analysis to **SI Sect. S2** and discuss it in the revised **Sect. 3.1.4** concisely. The authors agree that the former Sect. 3.4 (ice residual analysis) contributes little to the overall outcome of this study. We decided to omit the discussion of ice residual from this manuscript. More sample analyses towards understanding molecular-level properties of soil dust ice crystal residual will be conducted for a separate publication.

RC: The use of "T" and "Ts" instead of "temperature" and "temperatures" add too much noise to the manuscript.

AR: All T and Ts are written out as temperature or temperatures in both the main manuscript and SI. The use of the abbreviation, *T*, is limited to the part of parameter expression (e.g., $n_{INP}(T)$ = INP concentration per unit standard air volume as a function of temperature), which is explained in the text.

RC: Lines 66-67: "(i.e., the freezing propensity of INP immersed in supercooled water)". Who freezes the INP or the droplet?

AR: It is the aerosol particle(s) immersed in a droplet. To increase the clarity, we rephrased the definition of immersion freezing as "the freezing of aerosol particle(s) immersed in a supercooled droplet".

RC: Lines 95-96: How about convection?

AR: Convection can certainly play a role, and it seems self-evident given the differential heating of a feedlot surface and surrounding vegetation. However, none of the authors, including experts in the OLLF research and agricultural engineering, are aware of any previous study showing it is the cause of vertical transport of OLLF dust.

In **SI Sect. S5**, the authors also added a note of "We note that our estimation of n_{INP} is limited at the source location. Further understanding of OLLF-derived INPs in the atmosphere will require future research in the dust generation mechanisms in association with local dynamics and thermodynamics, vertical distribution of OLLF dust, and their fate in the atmosphere".

RC: Line 99: "where a convective cloud and updraft system persists". What does it mean? that you have such a system 365 days per year?

AR: We admit that "persistent" is not the right word. We meant to say that we frequently observe such systems in this region. We decided to rephrase this sentence to "Convection and updraft system may also help the vertical transport of aerosol particles in the Southern High Plains region (Li et al., 2017).".

RC: Lines 115-118: This is not the right place. Please add this text earlier.

AR: Moved to the 4th paragraph in the Introduction section, where we first mention "manure".

RC: Section 2.2. This Section is not easy to follow. It is longer than needed and it has to be rewritten to be more concise and clear.

AR: We moved sample analysis outcomes to the Results and Discussion section and leaving only a concise technical description in the Materials and Methods section. To improve the overall conciseness and readability of the methods section, the authors reorganized the section based on (2.1) laboratory study and (2.2) field investigation.

RC: Lines 154-155: Add the depth and how were they collected?

AR: We have clarified this information in the revised **Sect. 2.1.1**:

"Soil samples were collected on September 20, 2017. All samples were scooped from the loose dry surface layer of the pens (< 5 cm). Typically, the pen surface layer only extends to a depth of about 5 cm, which represents the depth of hoof penetration into the pen surface (Guo et al., 2011). This surface layer is rich in loose manure, which is a major source of ambient OLLF dust (Bush et al., 2014; von Holdt et al., 2021). All samples were ground and sieved for grain size < 75 μm. They were kept in chemically inert containers at room temperature until analyzed".

RC: Line 186: How about wind speed?

AR: Addressed above.

RC: Line 345: "Even assuming we evaluate INP up to 2,000 L-1, our INP fraction is 1%. Thus, our ns parameterization is reasonable." I don't get the message here.
AR: We reclarified this in the revised **Sect. 2.1.3**. The INSEKT system typically measures INP counts up to several hundred. For this study, the highest INP concentration measured by INSEKT was 135.9 INP L^{-1} (95% confidence intervals, CI95% = 80.1 – 198.5 L^{-1}) from the TXDUST01_08 experiment. As seen in **Table 1**, for this particular experiment, the filter sampling activity for INSEKT was conducted with an aerosol concentration of 266.3 x 10^3 L^{-1}. This simply translates to INP fraction of < 1%, which satisfies the prerequisite of n_s application addressed in Niemand et al. (2102).

RC: Line2 354, 374 and 375: "cumulative mass", "cumulative PM mass", and "aerosol particle mass". What does it mean? In methods two different systems to measure PM10 were described. Do the authors refer to PM10?
AR: Discussed above – please see Table 2 in this author response document.

RC: Lines 363-364: "However, because the measured nINP is low at high T, the CI95% error of nINP,upwind at around -15 °C is relatively large as compared to that at a lower T (Fig. 3a)". I do not get it.
AR: For clarity, we rephrased it to "… At this temperature, the $n_{\mathrm{INP,upwind}}$ (CI95%) error in a log scale spectrum is relatively large as compared to the lower temperature region, and the difference between $n_{\mathrm{INP,downwind}}$ and $n_{\mathrm{INP,upwind}}$ is not conclusive beyond the uncertainty around -15 °C".

RC: Line 366: "on local meteorological conditions". What was the typical wind speed?
AR: The measured wind speed was on average ≈ 10 miles per hour (min to max = 3.6 to 23.3 miles per hour). As explained above, the wind seems not to have much to do with the resuspension of feedlot surface materials, but certainly could contribute to instantaneous spikes of PM_{10} as our field sampling activities were carried out in the proximity of livestock pens (**Sects. 2.2.2 and 3.2.1**).

RC: Line 367: "short episode of soil dust". What do the authors mean? Resuspended dust?
AR: Yes. We replaced it with "resuspended OLLF soil dust".

RC: Lines 388.389: "This motivates the need for further characterization of our OLLF samples in a controlled-lab setting in order to identify what particulate size population (i.e., supermicron vs. submicron) and other properties trigger their". This cannot be evaluated in the field? should not just simply change the cut-off of the filter sampling PM1.0 vs. PM10?
AR: It could have been. But we did not have proper apparatus during our field investigation and, thereby, did not and perform PM_1 and PM_{10} sampling in our fields. Nevertheless, we show there is a notable correlation between INP and PM_{10} based on our 2017 – 2019 field study, which indicates the importance of large supermicron aerosol particles as INPs. This result supports the DFPC characterization of our OLLF samples in a controlled-lab setting to identify what particulate size population (i.e., supermicron vs. submicron). The onsite measurements of size-segregated INPs with a combination of a size-selecting impactor inlet and an online INP monitor will be indeed meaningful to add insights on the importance of large INPs. We include this point in our conclusion.

RC: Lines 399-410: This belongs to methods. Most of this information is already known from previous experiments in the AIDA, and therefore, their contribution to the manuscript is little and should not be in this section.
AR: The authors would like to keep this information here. We consider this as specific 'results' during our TXDUST01 campaign. We have changed the sub-section tile to "**3.1.2. AIDA measurements and freezing efficiencies of surface materials**".

RC: Lines 420-421: "the INSEKT results suggest that the bulk TXD01 sample is more active than filter-collected samples". Is this not expected as the bulk likely contain larger particles?

AR: The authors are not sure if it could be expected or not. Previously, Boose et al. (2016) studied immersion freezing abilities of diverse natural dust samples from nine desert regions around the globe (4 airborne and 11 sieved/milled surface samples) and found that the surface-collected samples tend to contain more efficient INPs than the airborne samples. The authors suggested that mineralogy may play a significant role to explain the observed difference. On the other hand, Kaufmann et al. (2016) found a similar freezing behavior of multiple surface dust samples despite the variation in mineralogy. Both studies noted the necessity of investigating non-mineral compositions (i.e., biological and other organics). While our laboratory and field samples are different in nature, our organic predominant samples show a reduction in IN efficiency for surface-collected samples compared to airborne field samples. The observed offset motivates further research in organic INPs. This point is now discussed in the revised **Sect. 3.3.1**. Note that we omitted the discussion regarding bulk vs. aerosolized but kept the immersion freezing efficiency of laboratory vs. field sample, scaled to the aerosol particle surface area (so the size factor is incorporated).

RC: Lines 424-425: "the lab-derived immersion spectra of both surface materials are reasonably comparable to the minimum – maximum boundaries of our field ns,geo spectra for T > -25 °C". If it is the same samples analyzed by different setups why should they show the large variability observed on the field samples?

AR: The authors found this part was misleading. We have rephrased it to:

"The immersion spectra of both surface materials are located towards the minimum boundaries of our field $n_{s,geo}$ spectra for temperature > -25 °C. While the variability of $n_{s,geo}$ at a single temperature could vary by several orders of magnitude for our field data, smaller variations are found for both lab results, implying different properties of our lab and field samples. The difference between our laboratory results and field data is discussed in **Sect. 3.3.1** in more detail".

An important caveat is that we could not find any notable inclusions of known IN-active microbiomes in both sample subsets. While we cannot rule out the possibility of IN from our field and laboratory samples triggered by biological INPs, our current results do not support it. The authors think that identifying heat-stable organic compounds and studying their physicochemical properties may be key to understand the properties of OLLF INPs. Our chemical composition analysis of laboratory samples (**SI Sect. S1**) indicates that they are exclusively organic in nature in terms of aerosol composition. Further, airborne particles collected in OLLFs are generally known to include substantial amounts of organic materials. For example, our previous work using Raman micro-spectroscopy revealed that ≈ 96% of ambient aerosol particles sampled at the downwind edge of an OLLF contain brown or black carbon, hydrophobic humic acid, water-soluble organics, less soluble fatty acids, and carbonaceous materials mixed with salts and minerals (Hiranuma et al., 2011). Recently, organic acids (i.e., long-chain fatty acids) and heat-stable organics were found to act as efficient INPs (DeMott et al., 2018; Perkins et al., 2020). However, our knowledge regarding what particular organics from OLLFs trigger immersion freezing at heterogeneous freezing temperatures is still lacking. A more detailed follow-up study to investigate molecular compositions of OLLF organics in ice crystal residuals may be necessary to provide an answer for it. This discussion is now given in the revised **Sect. 3.3.1**.

RC: Lines 424-426: While the variability of ns,geo at a single T could vary several orders of magnitude, similar variations are found for both lab and field results, implying the similarity of freezing efficiencies of our lab and field samples". This is expected for field samples as they are not identical and may have different composition, but in the laboratory, the instruments collected exactly the same samples.

AR: Addressed above – RE: RC: Lines 417-418 and Line 524. As shown in **Fig. 4**, the laboratory-measured $n_{s,geo}$ values are in agreement for the overlapping temperatures as all measurements are made using the same aerosol particles in the AIDA chamber. An offset between AIDA and INSEKT at around -22 °C may derive from the online vs. offline instruments issue, which is previously reported by some of the authors (Hiranuma et al., 2015). This is beyond the scope of the current study, and we would like to avoid extensive discussion about this issue.

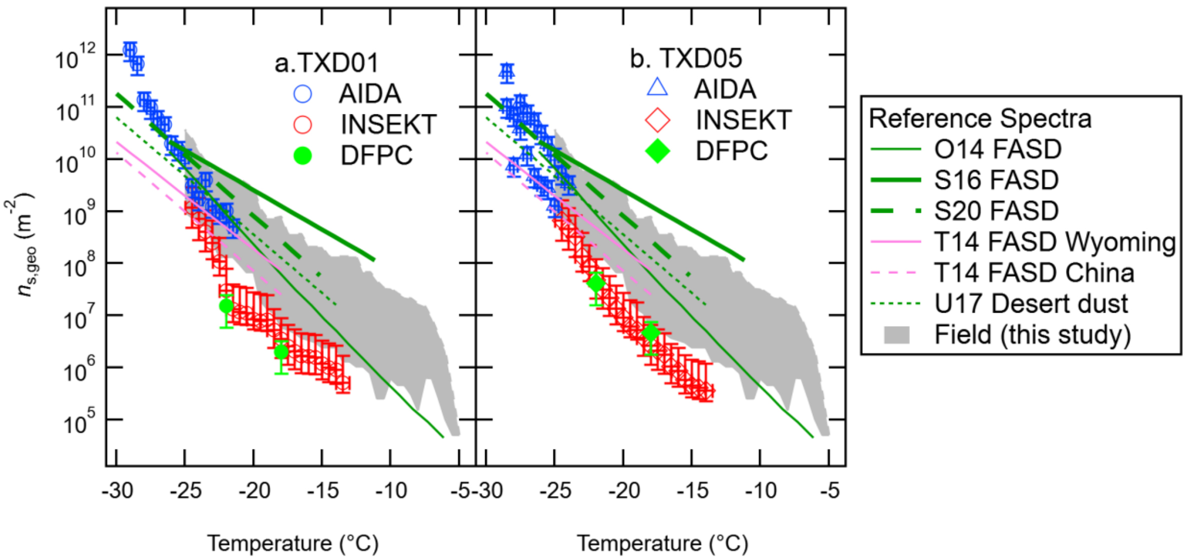

Figure 4. IN-active surface-site density, $n_{s,geo}$, of surface materials, TXD01 (a) andTXD05 (b), was assessed by AIDA, INSEKT, and DFPC (total aerosol particles) as a function of temperature. Six reference $n_{s,geo}$ curves for fertile and agricultural soil dust (FASD) and desert dust are adapted from O'Sullivan et al. (2014; O14), Steinke et al. (2016; S16), Steinke et al. (2020; S20), Ullrich et al. (2017; U17), and Tobo et al. (2014; T14). The grey-shaded area represents the range of our field $n_{s,geo}$ values at 0.5 °C interval for -5 °C > temperature > -25 °C (**Fig. 8**).

RC: Lines 427-430: "there is a difference in the INP abundance between bulk (< 75 µm-sieved) and aerosolized/filtered-samples for TXD01 (≨ 6.5 µm; Table 3) presumably due to different properties in particles of these two size subsets (6.5 – 75 µm and ≨ 6.5 µm) and/or different amount of IN-active soil organic matter". Again, is this not expected?

AR: Addressed above. The discussion regarding bulk vs. aerosolized samples has been excluded.

RC: Line 431: "be more representative of atmospherically relevant dust". Based on what? This needs to be clearly discussed.

AR: The authors agree that it is confusing. We excluded this ambiguous statement and rephrazed the sentence to:

"Additionally, the similarity of our lab results between TXD01 and TXD05 suggests that different physicochemical properties found for our samples may not impact their INP propensities".

RC: Line 433: "This comparability suggests that freezing ability is similar for condensation and immersion for our surface samples". I am not sure if such a strong conclusion can be said from just 2 data points from the DFPC.

AR: The DFPC measurements were carried out within the optimal operating conditions of the DFPC chamber. We understand the referee's concern. We have excluded this sentence from the Conclusions section and rephrased this part as:

"Moreover, the importance of large aerosol particles on immersion freezing was verified in our AIDA-based laboratory study. The DFPC offline freezing instrument assessed IN abilities of OLLF dust surrogates with PM_1 and total (> PM_1) size fractions. Our assessment revealed that on average ≈ 50% of OLLF n_{INP} derived from supermicron aerosol particle population in the assessed temperature range between -18 and -22 °C. Thus, our laboratory study showed the potential importance of supermicron aerosol particles from OLLFs as INPs. While our metagenomics analysis does not support the presence of known IN-active microbiomes, more research should be directed to reveal the compositional identities and associated IN abilities of various other animal feeding facility samples".

We have moved the associated discussion to the Results and Discussion section (**Sect. 3.1.3**):

"Besides, several unique characteristics of OLLF INPs were disclosed. For instance, comparability of results from our condensation freezing instrument (DFPC) and immersion freezing assay (INSEKT) was found for both sample types at the overlapped temperatures (18°C and -22°C). A similar observation was previously made for kaolinite particles in Wex et al.(2014). However, as the examined temperatures in our study are limited, the observed equivalence between immersion and condensation freezing for our surface OLLF samples should be cautiously interpreted and may not be conclusive".

RC: Line 569: "Our lab and field measurements-based parameterizations". It is not clear how the laboratory results were incorporated or used into the parametrizations. As stated in Major point #5 Lab and Field data should not be combined.

AR: Laboratory and field data were fitted independently (not combined). Please see our revised **Table S3**. We now only offer two lab parameterizations (for two laboratory samples, TXD01 and TXD05) and one field parametrization. In addition, the figure below shows our parametrization fits for those lab and field data.

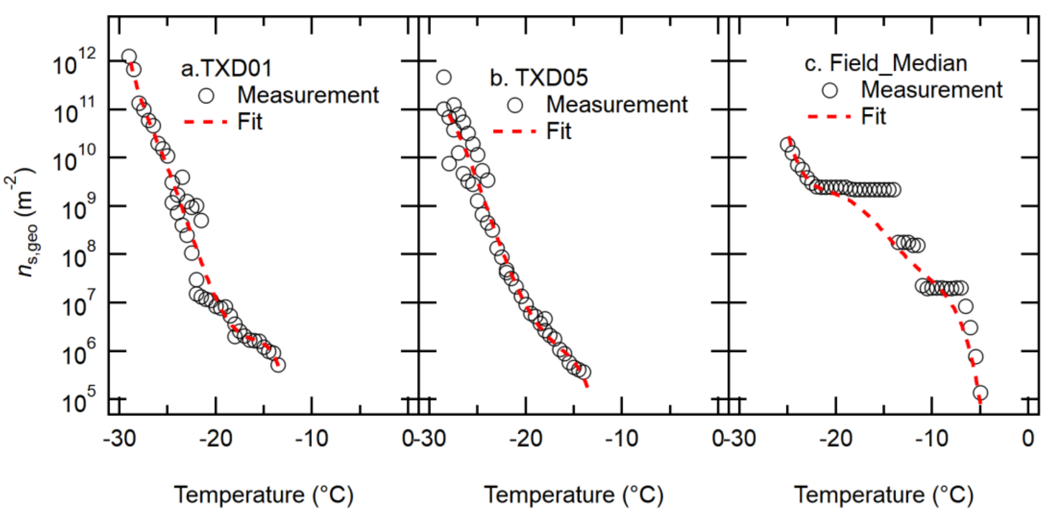

Figure. OLLF-INP parameterizations and fit curves based on Table S3 compared to our measurements for (a) TXD01, (b) TXD02, and (c) Field_Median.

RC: Lines 643-544: "Additionally, the observed consistency in the spectral slopes (i.e., Table 5) suggests that lab and field measurements exhibit similar IN ability at examined Ts". This was true at -25C not for the whole temperature range.

AR: The referee is right. There is a slight difference between the Δlog ($n_{s,geo}$)/ΔT values of laboratory results (~0.4) and that of the field (~0.5). The said sentence has been excluded from the manuscript. In general, our numbers are higher than what has been found in previous soil dust ice nucleation studies. This is now mentioned in the revised **Sect. 3.3.2**.
"Overall, the range of spectral slope deviations (0.41– 0.52) is higher than what we previously studied in soil dust samples in **Fig. 4** (0.15 – 0.27; S16 – O14), indicating a unique feature of the OLLF dust"

RC: Figure 5: In x-axis is it micro or milligrams? In Table 1 it is reported in micrograms
AR: Thank you for catching this. A "microgram" is right. The figure x-axis caption is corrected.

RC: Figure 7: it does not make sense to have a, b,c,d on each panel. The description of panels d are unclear. What is the concentration reported in red and blue?
AR: The panels (d) show two variables: (left axis) the number concentration of > 20 µm volume equivalent diameter particles measured by a welas optical particle counter, which is virtually equivalent to the number concentration of ice crystals measured during the AIDA expansion experiments; (right axis) the number concentration of aerosol particles in the AIDA chamber, measured by a condensation particle counter. It was clarified in L403-404 (now in the revised **Sect. 3.1.2**). We updated the figure axis texts accordingly. The authors would like to retain these panel IDs. We believe that retaining (a)-(d) offers a simple interpretation of panels.

RC: Figure 9: Why is it that noise? What is the time resolution? Would it not be better to do at a lower time resolution to avoid the noise?
AR: Time-resolution is 5-min. As part of our tapered-element oscillating microbalance (TEOM) data screening and evaluation protocol, all systematic errors (i.e., mass concentration outside of measurable limits, noise > 100%, 3.5 < main flow < 2.5, and 14 < sheath flow < 13) were excluded for our data analysis. The screened TEOM data were used as ambient particle emission data to estimate INP concentration from a feedlot. As stated in our manuscript, the resuspension/emissions mechanism of feedlot soil dust is not controllable as it highly depends on a unit of mobile livestock, which can be impulsive. Thus, these spikes are realistic (not any systematic errors). While time-averaging the data may eliminate some spikes in this figure, we would like to report processed individual data points in this figure. Please know that we offer seasonal time-averaged data of estimated n_{INP} in **Table S2**.

Technical comments:

RC: Lines 28: what is the meaning of "3 × 10-7 g L-1" .
AR: It is the minimum TEOM-measured aerosol particle mass concentration. The sentence, which included this number, is now excluded as the relevant discussion is moved to SI.

RC: Lines 48-51: Delete them.
AR: Deleted.

RC: Line 54: delete "chapter 9".
AR: Deleted.

RC: Line 56: Add other references in addition to Storelvmo (2017)
AR: Bourcher et al. (2013) and Zelinka et al. (2020) added.

RC: Line 78: "Agricultural land use is in excess of 50% of total U.S. land use". Please rewrite it.

AR: Corrected to:
"Agricultural land use accounts for more than 50% of total U.S. land use according to the U.S. Department of Agriculture (Bigelow and Borchers, 2012),…"

RC: Line 87: Add references after "conditions".
AR: We added Auverman (2001) and Postoor et al. (2012).

RC: Line 90: Add references after "head".

AR: We added Annamalai et al. (2012) and USDA (2021).

RC: Line 101: "we examinedthe". Fix it.
AR: Fixed.

RC: Line 106: Add references after "materials".
AR: This sentence has been removed. So no reference is provided in the revised manuscript, but a relevant reference could have included: National Research Council (NRC): Air Emissions from Animal Feeding Operations: Current Knowledge, Future Needs, Ad Hoc Committee on Air Emissions from Animal Feeding Operations, Committee on Animal Nutrition, NRC, 2003.

RC: Line 125: Add references after "definition".
AR: We now provide the U.S. EPA's URL - https://www3.epa.gov/npdes/pubs/sector_table.pdf

RC: Line 161: "using an offline freezing technique". Which one?
AR: INSEKT

RC: Line 171: "those of previously measured". Fix it.
AR: The measured BET specific surface area (*SSA*) values of OLLF samples are slightly higher compared to those of previously measured agricultural soil dust samples (0.74 – 2.31 m^2 g^{-1}; O'Sullivan et al., 2014),
→
The measured BET *SSA* values of OLLF samples are slightly higher compared to previously measured agricultural soil dust samples (0.74 – 2.31 m^2 g^{-1}; O'Sullivan et al., 2014),

RC: Line 184: Remove "Fig 5."
AR: Removed.

RC: Lines 196-198: "proxies. We chose the AIDA chamber as our study platform because it simulates ice formation in mixed-phase clouds in a controlled setting with respect to both T (± 0.3 °C) and humidity (± 5%; Fahey et al. 2014)." Delete.
AR: Deleted.

RC: Lines 201-203: "experiment. The AIDA has been applied for the analysis of both ambient and lab-generated INPs and has facilitated characterization of many INP species with the IN efficiency uncertainty of ± 39% (Steinke et al., 2020; Ullrich et al., 2017; Niemand et al., 2012; Hoose and Möhler, 2012)." Delete.
AR: Deleted.

RC: Lines 223-225: "Another motivation for using the AIDA facility is its ice-selecting pumped counterflow

virtual impactor (IS-PCVI; Hiranuma et al., 2016). As detailed in Supplemental Information (SI) Sect. S1, IS-PCVI separates ICRs from interstitial particles, including cloud droplets, at Ts below -20 °C." Delete.
AR: Deleted.

RC: Line 225: "evaporation". Should it be sublimation?
AR: Could be both. The authors incorporated sublimation in the main text.

RC: Line 300: "Texas dust". Delete.
AR: Done.

RC: Line 310: "Next, our metagenomics analysis method of total DNA is described". Delete.
AR: Deleted.

RC: Line 386: "at below -20 °C". Fix it.
AR: below -20 °C.

RC: Lines 386-387: "ambient aerosol particle mass concentrations based". PM10?
AR: Yes.

RC: Line 389: "in a controlled lab setting". Delete.
AR: Deleted.

RC: Line 412: "O14, S16, S20". Add the origin/source of the samples.
AR: The authors added the followings: O14 (England), S16 (Mongolia, Argentina, and Germany), S20 (Northwestern Germany, Wyoming), T14 (Wyoming), T14 (China), and U17 (desert dust samples from Aisa, Canary Island, Israel, and Sahara).

RC: Line 472: Add references after "mass".
AR: The authors added Hoose et al. (2010) in **Sect. 3.1.4**.

RC: Lines 486-487: "properties. All of our single particle analyses were carried out with the following parameters: electron beam accelerating voltages of 15 keV, spot size of 50, and working distance of 10 mm". This belongs to Methods.
AR: Ok, but this part is now omitted.

RC: Line 525: "We elected to use the". Fix it.
AR: Fixed.

RC: Line 529: "typically substantially lower". Fix it.
AR: Fixed.

RC: Line 580: "atmospherically relevant". What do the authors mean?
AR: The authors realize that this is too ambitious to retain, so deleted it. The authors understand that it requires more investigation to state it this way.

RC: Line 593: "dust samplew". Fix it.
AR: Fixed.

RC: Table 6. Last column "Spermicron Size". Fix it.
AR: Fixed.

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Response to Referee #4

The authors would like to express our sincere gratitude for the referee and helpful comments. Below, we provide our point-by-point responses. The referee's comments (RC) are shown from here on in black. The authors' responses (AR) are in blue below each of the referee's statements. We introduce the revised materials in green color along/below each one of your responses.

RC: The reviewed manuscript presents IN measurements and particle characteristics from particles emitted from select feedlot sites in the Texas Panhandle. The field measurements are complemented by laboratory measurements and the study benefits from the various analyzed particle characteristics. However, I would strongly suggest the authors to further revise the manuscript for conciseness and thus increasing readability and clarity of most sections, but particularly the methods and results sections.
AR: The authors appreciate these general remarks. Following the referee's advice, we revised our manuscript structure and contents to improve the readability and conciseness of this paper. Below, we provide our point-by-point responses.

RC: Some additional comments for the authors:
- Line 214/215: What is the scientific motivation of point (2)?
AR: Our motivation is to complement the aerosol interaction and dynamics in the atmosphere (AIDA) immersion freezing data at relatively high temperatures. The authors now clarified our point is **Sect. 2.1.3** as:
"The INSEKT data are especially useful to complement the AIDA chamber immersion results at temperatures above -25 °C".

RC: - Line 218/219: INSEKT covers a different size range than AIDA? If this is the main point for using INSEKT then the authors should mention this here to make it more clear why certain methods are used alongside others.
AR: No. We consider that both AIDA and ice nucleation spectrometer of the Karlsruhe Institute of Technology (INSEKT) cover the same aerosol particle size range as the aerosol particles for INSEKT analysis were directly sampled from the AIDA chamber. The sampler for INSEKT employed a sampling flow rate of 10 L min^{-1} to minimize in-line particle losses. For the ice nucleation efficiency estimation, both AIDA and ISEKT scaled ice-nucleating particle (INP) concentration to the same aerosol particle measurements (summarized in **Table 1**). We have added the following parts in our INSEKT methodology sub-section (**Sect. 2.1.3**) to clarify this point:
"…and S_{total}/M_{total} is a geometric specific surface area. The S_{total}/M_{total} value used for this study was derived from particle size distribution measurements from the AIDA chamber (presented in **Table 1**)".

RC: I would recommend to generally revise the methods section for conciseness and thus clarity with a focus on providing the reader with a clear overview of how methods complement each other.
AR: The authors agree with the referee. We realized that the structure of the method section and presenting various things could be overwhelming. The authors also acknowledge that retaining the format from a previous version would cause additional confusion. Therefore, the authors decided to restructure the main article to present the most invaluable scientific outcomes and limit the amount of Supplemental Information (SI) for the sake of readability. Our decision is based on considering all four referees' comments.

The authors revised the manuscript to feature the **abundance of supermicron aerosol particles acting as feedlot INPs from lab and field studies** and did the following modifications:

(1) Separating **Sects. 2 and 3** based on laboratory study (sub-section 1) and field investigation (sub-section 2) to explain methods, materials, and results for each sub-section independently.
(2) Moving sample descriptions to the Results and Discussion section (**Sect. 3.1.1**) and leaving only concise technique explanation in the Materials and Methods section (**Sect. 2.1.1**) to increase the readability of the manuscript in an organized manner.
(3) Moving the heat treatment data, outcomes, and discussion from the main manuscript into a single **SI Sect. S4**. Keeping it over different sections in methods and results impaired the overall readability in our previous version, and the authors believe that this modification resolves the readability issue.
(4) Removing all bulk sample discussions from this manuscript and focusing on the filter-collected aerosolized samples – So there will be no bulk vs. aerosolized sample discussion. In the end, the bulk is not our main outcome. The revised paper focuses on lab vs. field, we believe that the aerosolized sample is more relevant to what is in the field than the bulk sample.
(5) Removing the ice residual composition discussion from the main manuscript. We do not have statistically valid aerosol particle composition data of our 'field' samples from this study. As referee 3 is concerned, we cannot conduct the comparison of laboratory and filed sample compositions, and the former Sect. 3.4 contributes little to the main text. The authors agree that the composition analysis is not the main focus of this manuscript, and decided to exclude this part from the manuscript.
(6) Moving the discussion regarding the estimated INP concentrations to **SI Sect. S5**.

In addition, the authors also changed the title of our manuscript to "Laboratory and field studies of ice-nucleating particles from open-lot livestock facilities in Texas", which better represents our research.

RC: - Method on DNA analysis: does sterilization remove DNA which might interfere with the DNA of interest here?
AR: The authors think it is not an issue. We made sure to clean the sampler itself and all fittings with volatile reagent alcohol well ahead of each sampling activity. Although sterilization may not completely remove DNA from filters and filter holders, assembly was done with great caution as to not contaminate filter holders and filters with non-sample DNA. Also, sterilization causes DNA fragmentation to small fragments which will not be amplifiable during metagenomics analysis.

RC: - Please ensure that acronyms are defined the first time they appear in the manuscript, this will help the reader to better follow along without having to search for definitions. E.g., definition of ICR in line 226 missing.
AR: Ice crystal residual (ICR) was defined in L113. We checked acronyms. We also provide a list of abbreviations in **SI Sect. S6**.

RC: - Figure 1: OLLF schematic is helpful but not very clear. Would suggest to adapt the shape and relative size of the study areas. Additionally, it is not clear from the map where the boundary between OLLF 2 and 3 is.
AR: It was intentionally made that way to protect the identity of commercial cattle feeders. The authors modified the figure to point out roughly where they are within county boundary lines but would not be able to provide any information beyond (e.g., exact coordinates). Please see the revised figure on the next page.

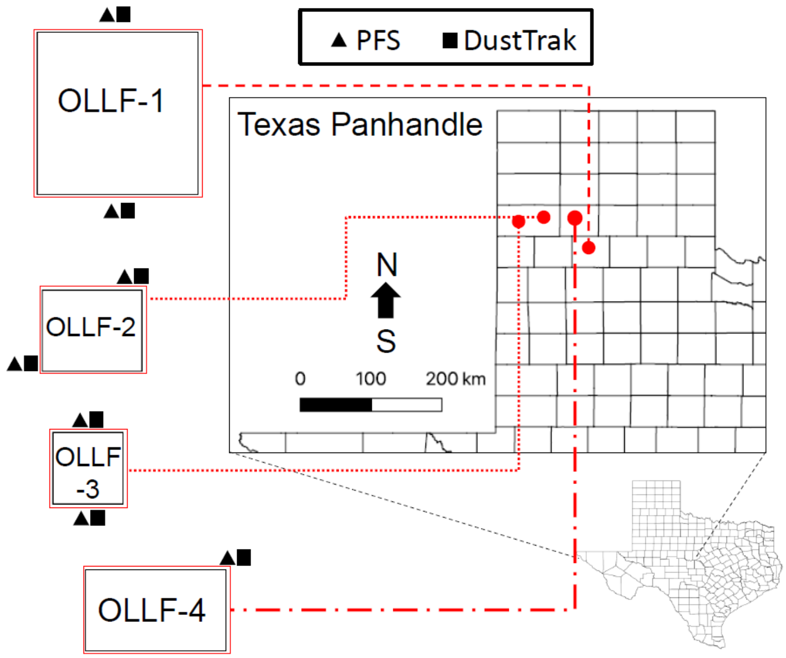

Figure 2. Schematic of the field sampling activity at individual sites (only the counties are shown). The dimension of each facility (east – west × north – south) is (1) 1.6 × 1.6 km, (2) 1.0 × 0.8 km, (3) 0.7 × 0.7 km, and (4) 0.8 × 1.4 km. A combination of polycarbonate filter samplers (PFSs) and DustTrak instruments was used at the nominally upwind and downwind edges of OLLF-1 to OLLF-3.

RC: - Comment 6. Ln 48 from reviewer 2, would suggest to also clarify in the manuscript
AR: To increase the readability and conciseness of the heat treatment part, all associated contents regarding heat treatment are now compiled in **SI Sect. S4**, and the clarification is provided in this section.

RC: - SI, particularly Figure S3: increase font size and/or image resolution for legibility.
AR: Done.

RC: - SI, Table S3: check for significant figures of the values provided in the table and be consistent.
AR: We now reduced significant figures as much as possible (please see the revised **Table S3**). We need to keep the reported significant figures to reproduce polynomial fits (We provide our fitting plots on the next page). It is susceptive to those decimals. As stated in the Table caption – "To reproduce the fitted curves, we needed to include all decimals". In addition, the authors have checked the consistency of significant figures and decimal digits for other tables, and we corrected them accordingly. For instance, in **Table 2**, we now report two decimal points. Air volume values and flow rates are scaled to the cross-section area of the filter examined.

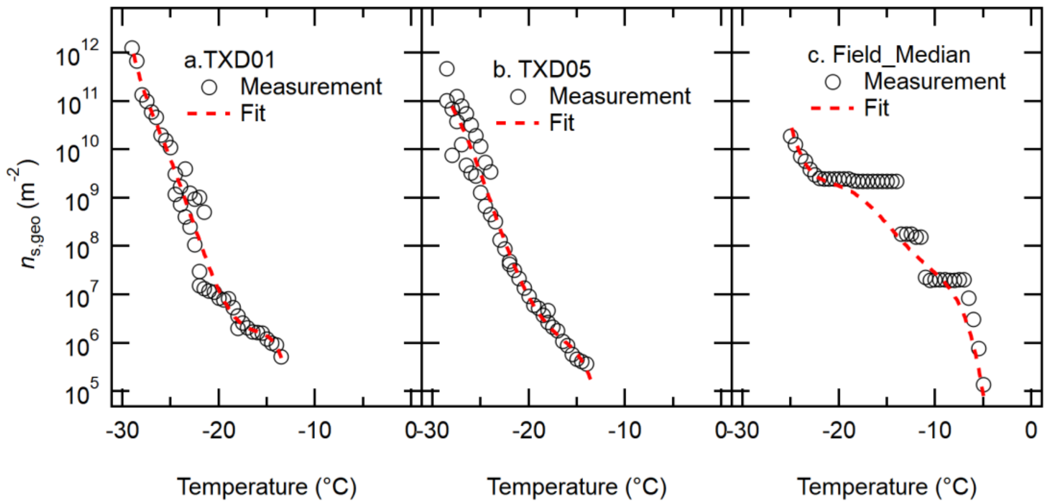

Figure. Open-lot livestock facility (OLLF)-INP parameterizations and fit curves based on Table S3 compared to our measurements for (a) TXD01, (b) TXD02, and (c) Field_Median.