

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 other 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). All references are available in the end of this AR document.

RC: This study reports the immersion ice nucleation ability of ambient feedlot aerosol samples and surface materials. Several instruments were engaged to perform physical, chemical and biological characterization, as well as immersion freezing ability determination of these filter samples and proxies. The authors have done great job characterizing and the presented data is abundant. However, the writing quality is pretty poor and the authors did not seem to offer a consistent usage of terms and symbols throughout the paper, which in part is hard to follow. More importantly, there are a few aspects need to be sufficiently addressed before this paper can be reconsidered for acceptance: First, the writing quality does require substantial improvement. Second, the paper structure is imbalance, with about 50% pages on materials and methods yet I still feel difficult to get the logic connection and setup of these experiments. Third, the Results and Discussion part is basically an experiment report with little analysis and discussion.

AR: The authors appreciate these general remarks and constructive criticisms regarding our manuscript by referee #1. We found these as invaluable guidance. We believe that the analysis and discussion in the revised manuscript are robust. We have very good data from our three-year field survey and the lab setting. However, some insufficient discussions might have led some of our data interpretation and presentation in an original manuscript to be unclear. The authors appreciate the peer-review comments, which motivated further analyses and improved the overall presentation. To allay the reviewer's concerns and mitigate any misgivings, the authors decided to change the title of manuscript to "Ice-nucleating particles from open-lot livestock facilities in Texas", reflecting our changes and articulate what is truly presented in the revised version paper. We have also revised our abstract, the conclusion section, and overall structure to reflect all of our major revisions and to increase the readability of this paper with rigorous analysis and discussion (please read the Track Changes version paper). Further, a consistency of terms and symbols has been checked. Below, we provide our point-by-point responses in hopes of our manuscript being considered for another review by the reviewer.

Major comments:

RC: 1. Introduction: The logic of this part needs to be thoroughly improved. A much more detailed review of relevant previous work should be added.

AR: The logic of this section has been improved by introducing (1) the climatic impact of ice-nucleating particles (INPs), (2) previous fertile-and-agricultural soil dust-derived INP studies (Suski et al., 2018; Conen et al., 2011; Hill et al., 2016; Steinke et al., 2016; 2020; Tobo et al., 2014; O'Sullivan et al., 2014 – more discussion along with our results in Sect. 3.6), and (3) potential significance of soil dust INPs in the U.S. as well as Texas (and the reasons) in the first four paragraphs.

RC: The following points should be explicitly addressed to give the audience an idea about the importance of this work: Why is feedlot dust relevant to mixed-phase clouds?

AR: Our research hypothesis and objectives are now also clarified in the introduction section as, “Due to the potential to act as a prevalent point source of microbiome-enriched dust particles in the Southern High Plains region, where a convective cloud and updraft system persists (Li et al., 2017), we hypothesized that an OLLF can be a source of soil dust INPs. To verify this hypothesis, IN propensities of aerosol particles from OLLFs, IN efficiencies of OLLF proxies, and their physicochemical and biological properties were studied in both field and laboratory settings.”

The authors also reformulated the third paragraph of our introduction section to point out the high loading of feedlot dust and potential impact of erosion by citing previous contributions and findings made by our team (i.e., Von Essen, S. G. and Auvermann, 2005; Bush et al., 2014; Hiranuma et al., 2011; Steinke et al., 2020) as follows; “In particular, the Texas Panhandle (northern most counties of Texas; also known as West Texas) is a major contributor to the U.S. cattle production, accounting for 42% of fed beef cattle in the U.S. and 30% of the total cattle population in Texas (> 11 million head). Annually, these cattle produce > 5 million tons of manure, which represents a complex microbial habitat containing bacteria and other microorganisms, on an as-collected basis (Von Essen and Auvermann, 2005). Agricultural dust particles observed at OLLFs have long been known to affect regional air quality because the dust emission flux and 24-hour averaged ground-level dust concentration can be as high as $23.5 \mu\text{g m}^{-2} \text{s}^{-1}$ and $1,200 \mu\text{g m}^{-3}$ (Bush et al., 2014; Hiranuma et al., 2011). Furthermore, our previous study revealed a presence of OLLF-derived particles at 3.5 km downwind of the facility, suggesting their ability to be transported regionally (Hiranuma et al., 2011). Moreover, some recent studies suggest that aerosol particles emitted from agricultural activities might reach cloud heights due to wind erosion, scouring, and other relevant mechanisms (Steinke et al., 2020 and references therein; Duniway et al., 2019; Katra, 2020).”

RC: How representative (e.g. size distribution, chemical composition, etc.) are the surface samples collected in this study compared to actual ambient aerosols?

AR: Composition: Organic particles with small salt contents (e.g., potassium) are predominantly present in surface-derived OLLF particle composition from this study, which is consistent with our previous study of ambient OLLF soil dust particle composition analyses (Hiranuma et al., 2011). Thus, in terms of composition, the authors believe that our surface-derived samples represent atmospherically relevant OLLF dust. Further, our study revealed a relative increase in organic inclusions (and decrease in salt inclusion) in ice crystal residuals, highlighting the importance of organic material for atmospheric immersion to be OLLF-derived INPs. Our previous work using Raman micro-spectroscopy revealed that ambient aerosol particles sampled at OLLFs contains brown or black carbon, hydrophobic humic acid, water soluble organics, less soluble fatty acids, and carbonaceous materials mixed with salts and minerals. But, our current knowledge regarding IN active organics is still limited. In the future, more detailed research should focus on understanding IN active organics common in soil dust. These important remarks are now stated in the new conclusion section (please see the track change version manuscript).

Size: As shown in Fig. 5 of Hiranuma et al. (2011), the surface area distribution of ambient OLLF dust measured with a combination of a GRIMM sequential mobility particle sizer and a GRIMM portable aerosol spectrometer peaks in a mode at $\sim 10 \mu\text{m}$ diameter. This mode diameter is somewhat larger than what we measured after aerosolizing our surface-derived samples in the AIDA chamber (summarized in Table 3). Note that we used cyclone impactors for the injection of OLLF surface sample into the AIDA to reduce the concentration of large aerosol particles to not misclassify them as ice crystals. But, it should be also 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. The difference mentioned above and the demand of controlled investigation were exactly our motivation of analyzing IN properties of both bulk samples ($< 75 \mu\text{m}$ -sieved) and aerosolized samples ($\approx 6.5 \mu\text{m}$). While we observed

a reasonable agreement between bulk and aerosolized IN propensity for the TXD05 sample within our experimental uncertainty is represented by 95% binomial confidence interval, there was a substantial difference of IN efficiency in between bulk and aerosolized one of the TXD01 sample. Overall, these results imply that TXD05 might be more atmospherically relevant. These points are now addressed in our new Sect. 3.2. To clarify our intention, we added the following sentence in the end of Sect. 2.2 – “As demonstrated in our previous study, the surface area distribution of ambient OLLF dust peaks in mode diameter at $\sim 10 \mu\text{m}$ (i.e., Fig. 5 of Hiranuma et al., 2011). This mode diameter is larger than surface-derived samples aerosolized and examined in the AIDA chamber (**Table 3**). 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. The difference mentioned above and the demand for controllable investigation motivated analyzing IN properties of both bulk samples ($< 75 \mu\text{m}$ -sieved) and aerosolized samples ($\approx 6.5 \mu\text{m}$). Further results and discussions about representativeness of the surface samples used in this study compared to ambient OLLF soil dust are provided in **Sect. 3**.”

RC: How representative are the sampling sites for the whole Texas?

AR: This is a valid question. We now clarify the representativeness of our field study sites in Sect. 2.1 as follows; “Four commercial OLLFs, ranging from 0.5 to 2.6 km² ($< 45,000$ head capacity), located in the Texas Panhandle region were used as the ambient aerosol particle sampling sites. All four sites are located within a 53 km radius of West Texas A&M University in Canyon, Texas. Our experimental layouts at each site, denoted as OLLF-1 to OLLF-4, are shown in **Fig. 1** (no further specification is provided to protect location privacy). All sites have a capacity greater than 1,000 head, which are categorized as large concentrated animal feeding operation facilities for cattle under the U.S. Environmental Protection Agency’s definition. These OLLFs were selected primarily for the east-west orientation of their feeding and working alleys, which were nearly orthogonal to prevailing south to southwest winds, allowing for downwind and upwind sampling. Our sampling sites represent typical OLLFs, as more than 75% of cattle are produced in large concentrated animal feeding operation facilities in the U.S. (Drouillard, 2018).”

RC: The title made a very broad and general statement without any solid support.

AR: The authors adapted a new title of “**Ice-nucleating particles from open-lot livestock facilities in Texas**” to reflect our changes and articulate what is truly presented in the revised version paper.

RC: 2. Materials and methods: The authors used 4 out of 9 pages to describe the materials and instruments used in this study. But it remains difficult to structure the experiments conducted. Please streamline this section and provide necessary experiment information.

AR: This is a valid point. The authors made a major revision in the manuscript structure, and the revised manuscript has the following structure and balanced page allocations; (1) Introduction: Page 1-2, (2) Methods: Page 3-6, (3) Results and Discussion: Page 6-10, and (4) Conclusion: Page 11.

RC: 3. As the main body of the manuscript, the results and discussion section is divided into 7 separate subsections without logical connection. Most of these subsection, simply report values and lack further analysis and discussion of the results. For instance, it is claimed that super-micron INP fraction contributed $49.7\% \pm 6.0\%$ of total INP for TXD samples at -18°C and -22°C (P7L20-22). However, no analysis or interpretation of this conclusion is presented. In Table 4 (P23), the change of super-micron INP fraction at two temperatures before and after the dry-heated treatment is different, and what could be the reason

for this deviation? Please revise this part in depth and add comparisons of the results obtained in this study to previous relevant work.

AR: First, it is a good suggestion to add comparisons of the results obtained in this study to previous relevant work. The authors added the following comparative discussion in Sect. 3.2. The authors think that having this discussion in this section gives a smooth transition and also provides a link to Sects. 3.3. and 3.4. So, we thank the referee for this invaluable suggestion. The authors note that there has not been much discussion of large soil dust organics and their contribution to atmospheric ice nucleation in previous studies. Tobo et al. (2014) mentions the need for analyzing soil dust INPs using mono- vs. poly-disperse particles, and the authors agree with the necessity of a systematic study of size-segregated soil dust INPs in the future (beyond the scope of this work). Hence, direct implication of what composition contributes to IN at different T s of OLLF particles is still not available. Further, while we did not see a systematic increase of supermicron INP fraction as a function T as shown in Mason et al. (2016; i.e., INP fraction at $-15\text{ }^{\circ}\text{C}$ > that of $-20\text{ }^{\circ}\text{C}$), our results in **Table 4** support that $n_{\text{INP,total}}$ is always higher than $n_{\text{INP,PM1}}$ for any type of samples used in this study. These points are now addressed as follows; “Since DeMott et al. (2010) successfully demonstrated the correlation between immersion-mode n_{INP} and the number concentration of aerosol particles larger than $0.5\text{ }\mu\text{m}$ diameter based on the compilation of field data for more than a decade, a number of studies have shown the evidence that supermicron aerosol particles dominate INPs across the world. For example, Mason et al. (2016) reported a substantial fraction of supermicron INPs through immersion freezing at relatively a high T (> 78% at $-15\text{ }^{\circ}\text{C}$) measured at seven different sites over North America and Europe. Even at $-20\text{ }^{\circ}\text{C}$, the author reported the fraction of supermicron INPs larger than 50%. Compared to these numbers, our laboratory data show lower fractions, but the INP sources are presumably different. Based on findings from recent study of size-resolved INPs vs. fluorescent biological particles, these INPs activated at $-15\text{ }^{\circ}\text{C}$ are typically thought to be biological (e.g., Huffman et al., 2013; Huang et al., 2021). While there has been more evidence that terrestrial and marine biological particles play an important role in immersion freezing of supermicron-sized particles (e.g., Ladino et al., 2019; Si et al., 2018; Creamean et al., 2018), the atmospheric implication of such rare aerosol species and the overall impact on aerosol-cloud interactions is still under debate. More recently, high IN efficiency by supermicron INPs derived from quartz-rich atmospheric mineral dusts have been reported from different locations, including East Asia (Chen et al., 2021) and eastern Mediterranean (Reicher et al., 2019). These mineral components usually contribute to IN at low T s. However, there has not been much discussion of large soil dust particles, especially organics, and their contribution to atmospheric ice nucleation in previous studies. Hence, direct implications of which components contribute to IN at different T s to the observed freezing properties of OLLF particles is still missing. Lastly, while we did not see a systematic increase of supermicron INP fraction as a function of T as shown in Mason et al. (2016; i.e., INP fraction at $-15\text{ }^{\circ}\text{C}$ larger than at $-20\text{ }^{\circ}\text{C}$), our results in **Table 4** support that $n_{\text{INP,total}}$ is always higher than $n_{\text{INP,PM1}}$ for any type of samples used in this study.”

Second, the letter-type manuscript content (remained for this particular section) is now revised to fit in the research manuscript format. The authors believe that the revised version provides sufficient analysis of the results and observations.

Specific comments:

RC: - P1L18: Please capitalize each word.

AR: Each keyword is now capitalized.

RC: - P1L21: Please add “the” before U.S. and check the usage of “the” throughout.

AR: Added and all consistent throughout now. The article usage in the main manuscript and SI has been re-checked with a native English speaker/PhD.

RC: - P1L28: “thereby finding...” is not clear to me.

AR: Rephrased to “leading to find...”.

RC: - P1L30: Please replace “improved” with “improving”.

AR: Replaced. Thank you.

RC: - P1L32: Please rephrase to “...explore the relationship between INP and supercooled cloud properties...”.

AR: Rephrased as suggested.

RC: - P1L38: 25% of total what?

AR: We meant total “total global dust emission” as stated in Sect. 7 of Ginoux et al. (2012)

RC: - P1L46: Please rephrase.

AR: We rephrased this sentence now in L89 as “Agricultural dust particles observed at OLLFs have long been known to affect regional air quality because the dust emission flux and 24-hour averaged ground-level dust concentration can be as high as $23.5 \mu\text{g m}^{-2} \text{s}^{-1}$ and $1,200 \mu\text{g m}^{-3}$ (Bush et al., 2014; Hiranuma et al., 2011).”

RC: - P1L48: “suggesting their regional scale impact” is not clear to me.

AR: For clarity, we rephrased these words to “suggesting their ability to be transported regionally”.

RC: - P1L50: Please verify the usage of “impact in”.

AR: It should have been “influence on”, but the authors decided to remove this sentence to improve the logical flow of this section with only the most relevant discussions and references.

RC: - P1L51: Please spell out “IN”, as this is the first appearance of this term in the text. Please check the whole paper for first appearance of abbreviations.

AR: Corrected. The authors apologize for missing abbreviation definitions in the main text. Admittedly, we overlooked the guideline given in the ACP web - “They need to be defined in the abstract and then again at the first instance in the rest of the text.”

RC: - P1L54: Please add the classical textbook of Pruppacher and Klett (2010). Please also spell out “INP”.

AR: Pruppacher and Klett (2010) is added, and INP is defined at its first appearance.

RC: - P1L56: Please rephrase “focused in this study”.

AR: The authors rephrased the sentence and now it read, “We focused on the immersion mode freezing because recent modeling simulation and remote sensing studies suggest that immersion freezing is the most prominent heterogeneous IN mechanism, accounting for 85 to 99%, through which ice crystals are formed in mixed-phase clouds (Hande and Hoose, 2017; Westbrook and Illingworth, 2011).”

RC: - P2L14: Please spell out “Ts” or specify the meaning of the subscript.

AR: Defined - temperatures (Ts).

RC: - P2L15: Please spell out “ICR”.

AR: Defined - ice crystal residual (ICR) samples.

RC: - P2L39: Please add “as” before addressed.

AR: Added as suggested.

RC: - P2L43: Please spell out “AIDA”.

AR: Defined.

RC: - P2L47: Please spell out “TXD”.

AR: Defined - Texas-Dust-01 (TXD01) and Texas-Dust-05 (TXD05)

RC: - P2L58: What instrument did the author use to measure BET SSA?

AR: Autosorb iQ model 7 gas sorption system (Anton Paar, former Quantachrome Instruments) – we rephrased the sentence as follows, “The Autosorb iQ model 7 gas sorption system (Anton Paar, former Quantachrome Instruments) was used to measure BET SSAs in this study.”

RC: - P3L1: Does the author imply that TXD05 is soil dust while TXD01 contains K-feldspar here? If so, why is the density of TXD05 larger than that of TXD01?

AR: No, the authors did not mean or imply an inclusion of K-feldspar. We simply meant that the increase in porosity can lead to higher BET SSA. To avoid any confusions, the authors decided to limit our BET SSA comparison to a previous soil dust study (O’Sullivan et al., 2014) and exclude two K-feldspar study references. Excluding these references does not change the story of this paper.

RC: - P3L3-8: Why did the author compare geometric and BET SSA directly and attribute the difference between these two totally different methods to sample particle size variance?

AR: Both surface parameters are important for ice nucleation active surface site density estimations – $n_{s,BET}$ and $n_{s,geo}$ (Hiranuma et al., 2015), which is cited within the relevant sentence. Since BET SSA is typically measured for ‘bulk’ sample, it may not represent the specific surface area of aerosolized particles, which was discussed in our previous study (Hiranuma et al., 2019).

RC: - P3L15: Please see the comment above.

AR: Pruppacher and Klett (2010) is added above. At the first appearance of immersion freezing (L54), we decided to give its definition (i.e., the freezing propensity of INP immersed in supercooled water) and cite relevant references of soil dust immersion freezing (Suski et al., 2018; Conen et al., 2011; Hill et al., 2016; Steinke et al., 2016).

RC: - P3L17: What is the purpose of filter sampling? Can the authors provide a scheme of the experimental setup in this study?

AR: These are all clarified in Sect. 2.3. The authors thank the referee for this suggestion to use experimental schematics. The authors believe that Fig. 2 clarifies and improves the readability of the section.

“An overall AIDA experimental schematic is shown in Fig. 2. Our OLLF dust proxy sample was injected into the AIDA chamber in an aerosolized form through a rotating brush disperser (PALAS, RGB1000) followed by passing through a series of inertial cyclone impactor stages to limit particle size to $< 10 \mu\text{m}$ in D_{ve} . Subsequently, the OLLF particle size distribution in the AIDA chamber was measured prior to each simulated adiabatic expansion experiment. Specifically, a combination of a scanning mobility particle sizer (SMPS, TSI Inc., Model 3080 differential mobility analyzer and Model 3010 condensation

particle counter), an aerosol particle sizer (APS, TSI Inc., Model 3321), and a condensation particle counter (CPC; TSI Inc., Model 3076) collectively measured the total number and size distribution of aerosol particles at a horizontally extended outlet of the AIDA chamber (Möhler et al., 2006). As seen in Fig. 2, a set of complementary filter samples of the aerosol particles directly from the AIDA chamber was also collected prior to expansion experiments for three purposes: (1) examining the condensation/immersion freezing ability of aerosol particle collected on nitrocellulose membrane filters (Millipore HABG04700, nominal porosity 0.45 μm) in the dynamic filter processing chamber (DFPC; Santachiara et al., 2010), (2) using them to perform measurements with the IN Spectrometer of the Karlsruhe Institute of Technology (INSEKT; Schiebel, 2017; Schneider et al., 2021), and (3) conducting metagenomics analyses to study biological components of the aerosolized samples. Afterwards, each particle type (i.e., TXD01 and TXD05) was individually examined for its immersion freezing ability during expansion experiments. To complement the AIDA chamber immersion results, INSEKT was used for aerosol particles collected on 47 mm Nuclepore filters (Whatman WHA10417012, pore size 0.2 μm) as well as for < 75 μm sieved-bulk samples collected. The DFPC technique was also used to measure the number concentration, ice-activated fraction, and nucleation efficiency of the INPs under different T conditions and for different particle sizes (i.e., PM_{10} vs. total) collected on filters. DNA sampling for metagenomics analysis to study biological components of the OLLF bulk samples was also conducted on aerosol particles collected on the Nuclepore filters through an independent inlet.”

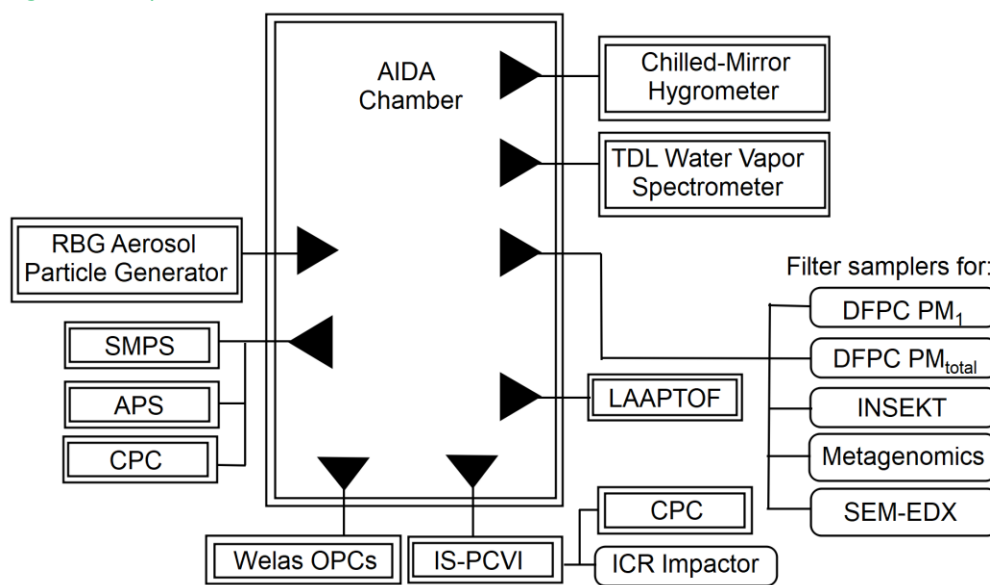


Figure 2. Lab experimental schematic of the AIDA facility. All samples were injected using a rotating brush generator (RBG) for aerosol particle generation. Multiple extramural instruments, welas optical particle counters (OPCs), an ice selective pumped counterflow virtual impactor (IS-PCVI), a hygrometer, a tunable diode laser (TDL) spectrometer, a laser ablation aerosol particle time-of-flight mass spectrometer (LAAPTOF; see SI), and aerosol particle counters/sizers (SMPS, APS, CPCs), are connected to the AIDA chamber. Downstream filters and an impactor collected aerosol particles and ice crystal residuals for multiple offline analyses.

In addition, we decided to provide a field experimental schematic as Fig. 1.

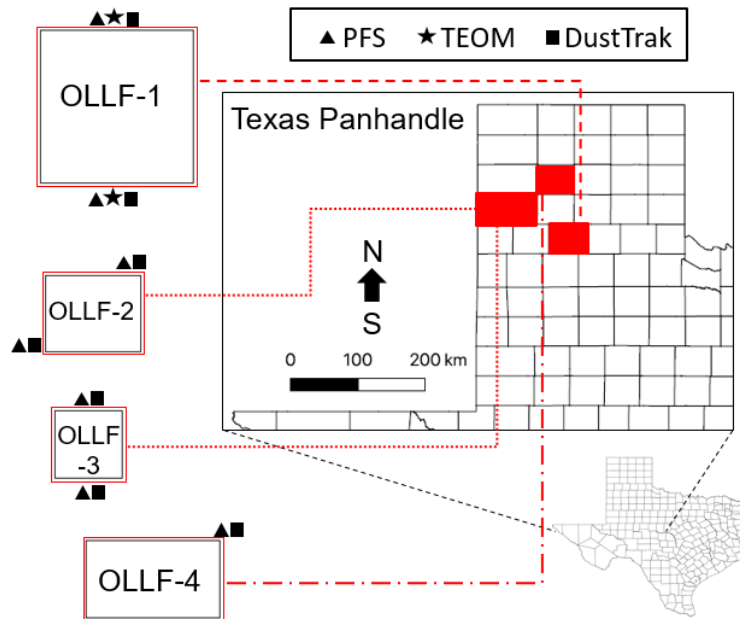


Figure 1. Schematic of the field sampling activity at individual sites. The dimension of each facility (east – west × north – south) is (1) 1.6 × 1.6 km, (2) 1.0 × 0.8 km, (3) 0.7 × 0.7 km, and (4) 0.8 × 1.4 km. A combination of polycarbonate filter samplers (PFSs) and DustTrak instruments was used at the nominally upwind and downwind edges of OLLF-1 to OLLF-3. Two tapered-element oscillating microbalances (TEOMs) were deployed at OLLF-1 alongside other instruments.

RC: - P3L24: Please add reference after “IS-PCVI”.

AR: Hiranuma et al. (2016) is now added.

RC: - P3L28: Should be μm here.

AR: Corrected.

RC: - P3L31: Does the authors mean “individual ice crystal” here? If so, maybe it’s better to delete “of individual particles”.

AR: We do not analyze the composition of ice crystals, we analyze leftover residual particles after evaporating all moisture. So the authors decided to reword it to “individual residual particles”

RC: - P3L41: Please add more AIDA references apart from the review paper.

AR: Steinke et al. (2020), Ullrich et al. (2017), and Niemand et al. (2012) are added.

RC: - P3L42-45: According to Table 3, the super-micron particle size can reach 5-6 μm . What’s the size detection limit of TSI SMPS and aerosol particle sizer?

AR: We used a combination of SMPS and APS to characterize the size distribution of particles injected into AIDA (addressed in Sect 2.3). The APS measures aerodynamic particle size from 0.5 to 20 μm . As stated in the manuscript, the SMPS and APS data were merged. The authors followed the method introduced in Möhler et al. (2006).

RC: - P3L45: What technique did the authors use to aerosolize particles?

AR: We used a rotating brush disperser (RGB 1000, Palas) as used in Möhler et al. (2006).

RC: - P4L2: Did the authors intend to say “array”?

AR: We meant “a set of”, and this part has been updated accordingly.

RC: - P4L7: Please spell out “HPLC”.

AR: Done - high-performance liquid chromatography (HPLC)

RC: - P4L8-9: How was the detection limit determined?

AR: Suspension water volume was optimized to assign first frozen droplet to correspond to 0.05 INP L⁻¹. A recently published paper describing the WT-CRAFT method (including the raised point) and its application (Vepuri et al., 2021) is added as a reference.

RC: - P4L11-12: Can the authors provide an example for such processing in SI?

AR: Our previous product provides a good example of visual separation of droplet-ice (Fig. 4 of Cory et al., 2019a; DOI: <https://doi.org/10.1002/essoar.10500739.1>). As other specifications of WT-CRAFT are sufficiently provided in Cory (2019b) and Vepuri et al. (2021), the authors would like to avoid including this in SI.

RC: - P4L25-26: If it’s “a series of diluted suspensions”, should it be “×15 to ×225” here? Besides, please replace “x” with “×” throughout the paper.

AR: All corrected in a consistent manner. Thank you.

RC: - P4L38: Remove the “of” after “100 L”.

AR: Removed.

RC: - P4L40: “was characterized”.

AR: Corrected.

RC: - P4L42: Please specify what type of “diameter”.

AR: Aerodynamic

RC: - Sect. 2.6: Please simplify the description of previous work.

AR: The authors believe that we need to retain all information in this section (now Sect. 2.5).

RC: - P5L31: What is the typical size range of droplets and ice crystals in this study?

AR: Droplet were typically < 24 μm and ice particles were > 24 μm optical diameter. For clarity, the figure shown below displays temporal evolution of droplet and ice crystal size distributions during the four ICR extraction experiments. Normalized size distributions of simulated cloud particles ($dN/d\log D_p$) were derived from the measurements in the welas optical particle counter installed below the AIDA chamber. As shown, a reasonable critical cut-size (> 24 μm) of the IS-PCVI was used for each expansion experiment.

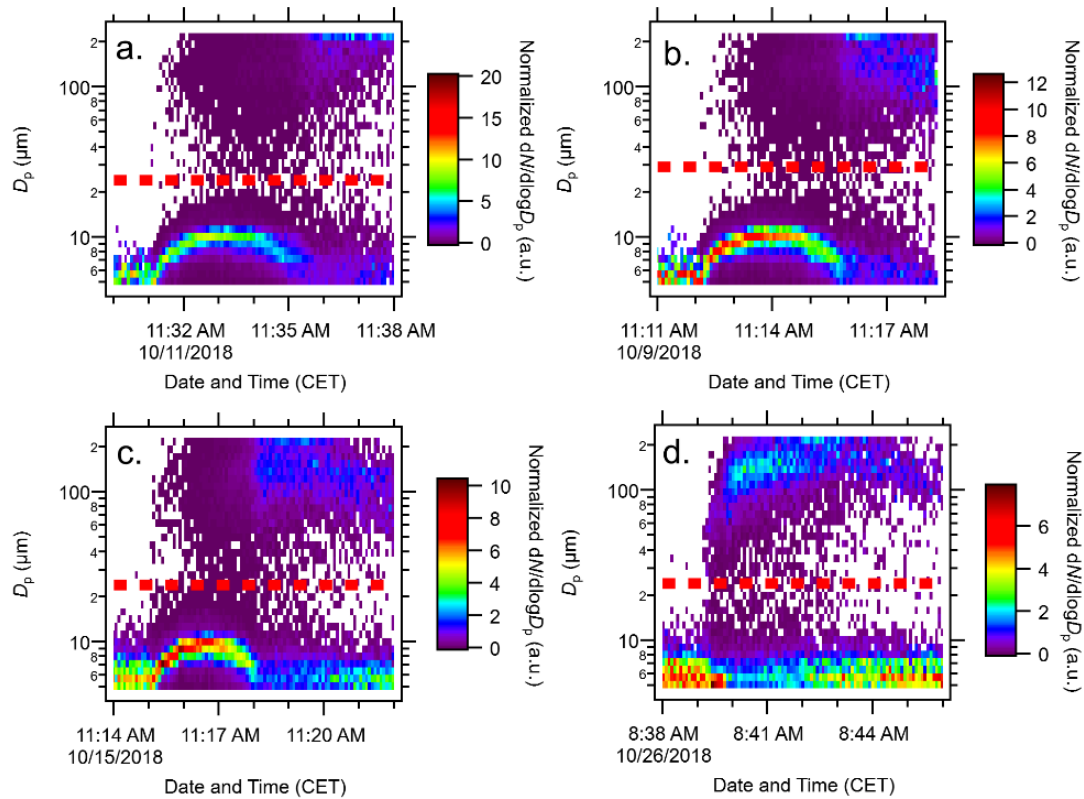


Figure. Temporal profiles of the weLas OPC size distribution of droplets and ice crystals in optical diameter (D_p) during individual expansions of TXDUST01_08 (a), _04 (b), _12 (c) and _31 (d). The red dotted lines represent the critical cut size, D_c .

RC: - P5L36: Should be μm . Please check the units throughout.

AR: Sorry. Checked and all corrected.

RC: - P5L37: During TXDUST01 what?

AR: We rephrased the sentence as: “During the TXDUST01 campaign, the output flow of IS-PCVI was constant at 2.5 lpm.” – it is now in SI Sect. S1.

RC: - P5L45: Please change the parenthesis to “(...Model 1400a; Patashnick...)”.

AR: Changed to “TEOMs; Thermo Scientific Inc., Model 1400ab; Patashnick and Rupprecht, 1991.”

Note: 1400ab is a correct Model no.

RC: - P5L58: Please enclose “ $<100 \mu\text{g m}^{-3}$ ” into parenthesis.

AR: Enclosed.

RC: - P6L3-9: What’s the point of repeating previous results in this paper?

AR: It was to clarify the observed supermicron ambient dust originates from a feedlot. As we now discuss this point using our new data, we decided to remove this repetitive discussion.

RC: - P6L11: I suggest to place the symbol right after the corresponding definition. Do the author mean INP concentration per unit particle mass and INP concentration per unit particle surface? Please specify and avoid misunderstanding.

AR: This is a good suggestion. Corrected as suggested.

RC: - P6L22: How were these quantities derived? Can the authors please provide the derivation or conversion process in SI apart from the references here?

AR:

The following is now available in SI Sect. S4 – derivation of n_{INP} , n_m , and $n_{s,geo}$:

S4. Derivation of n_{INP} , n_m , and $n_{s,geo}$

Here we describe the conversion procedure used to derive ambient n_{INP} , n_m , and $n_{s,geo}$. Initially, we computed the $C_{INP}(T)$ value, which is the nucleus concentration in ultrapure water suspension (L^{-1} water) at a given T as described in Vali (1971). This $C_{INP}(T)$ value was calculated as a function of unfrozen fraction, $f_{unfrozen}(T)$ (i.e., the ratio of number of droplets unfrozen to the total number of droplets) as:

$$C_{INP}(T) = - \frac{\ln(f_{unfrozen}(T))}{V_d} \quad [1]$$

in which, V_d is the volume of the droplet ($3 \mu L$) for WT-CRAFT and sample in a well ($50 \mu L$) for INSEKT.

Next, we converted $C_{INP}(T)$ to $n_{INP}(T)$: INP in the unit volume of atmospheric air at standard T and pressure (STP) conditions, which is 273.15 K and 1013 mbar. The cumulative n_{INP} per unit volume of sample air, described in the previous study DeMott et al. (2017), was then estimated as:

$$n_{INP}(T) = C_{INP}(T) \times (DF) \times \frac{V_l}{V_{air}} \quad [2]$$

where DF is a serial dilution factor (e.g., DF = 1 or 10 or 100 and so on). The sampled air volume (V_{air}) is given in **Table 1**. The suspension volume (V_l) is optimized to achieve the detection limit of 0.05 INP L^{-1} (corresponding to the first frozen droplet).

Finally, based on Eqn. 3 of Hiranuma et al. (2015), the $n_{s,geo}(T)$ and $n_m(T)$ values can be derived as:

$$n_m(T) = \frac{n_{INP}(T)}{M_{ve}} \approx \left(\frac{S_{total}}{M_{total}} \right) n_{s,geo}(T) \quad [3]$$

where M_{ve} is the mass of a spherical particle of volume equivalent diameter (g), and S_{total}/M_{total} is a geometric specific surface area (Hiranuma et al., 2015). The value used for converting field $n_m(T)$ to $n_{s,geo}(T)$ data, $\sim 0.4 \text{ m}^2 \text{ g}^{-1}$, is derived from particle size distribution measurements presented in Fig. 3 of Hiranuma et al. (2011).

RC: - P6L23: Please verify the usage of “Regardless”.

AR: Done. For clarity, the authors now simplify this sentence by combining with previous sentence as; “While the background freezing contribution of the field blank filter was negligible (< 3%) at -25 °C, we purposely limited our WT-CRAFT data analysis to the T range between 0 °C and -25 °C to eliminate any possible artifacts in our WT-CRAFT data.”

RC: - P6L25: How did the author decide whether the data was “uncertain systematically erroneous data”?

AR: This just means we are not confident with our WT-CRAFT data below -25 °C as artifacts become non-negligible (Vepuri et al., 2021), and we report our WT-CRAFT data only in the T range above -25 °C.

We have rephrased it to: to eliminate any possible artifacts in our WT-CRAFT data

RC: - P6L27-28: The difference between field samples and lab samples in Fig. 3(a) can reach up to three orders of magnitude between -20 to -10 °C, which is not “comparable” and is not capable of “validating” from my point of view.

AR: This is a valid question. The authors took the referee’s word as a motivation of further analyzing our data for spatial variability (Fig. 3) and seasonal variability (Figs. 4-6) to explain the observed data variation and completely reformulated Sect. 3.1. Please see the track changes manuscript.

RC: - P6L31: Is this linear relationship a linear correlation analysis or a linear regression fit? Please specify, and give the corresponding correlation coefficient and/or other parameters to indicate the goodness of this correlation.

AR: The authors apologize for extending the assessment to the raised point. The following description now appears in the Fig. 5 caption.

- Yes. It is a linear regression curve in log scale ($n_{\text{INP}} = 3.51 \times \text{Cumulative PM Mass} - 2.41$; $r = 0.94$).
- This is the value of representative n_m at the given T ($3.55 \times 10^9 \text{ g}^{-1}$), which is a median n_m value of minimum to maximum in order to guide the reader’s eye and to show an example of quasi-constant n_m of cattle feedlot-derived INP’s nucleation efficiency.

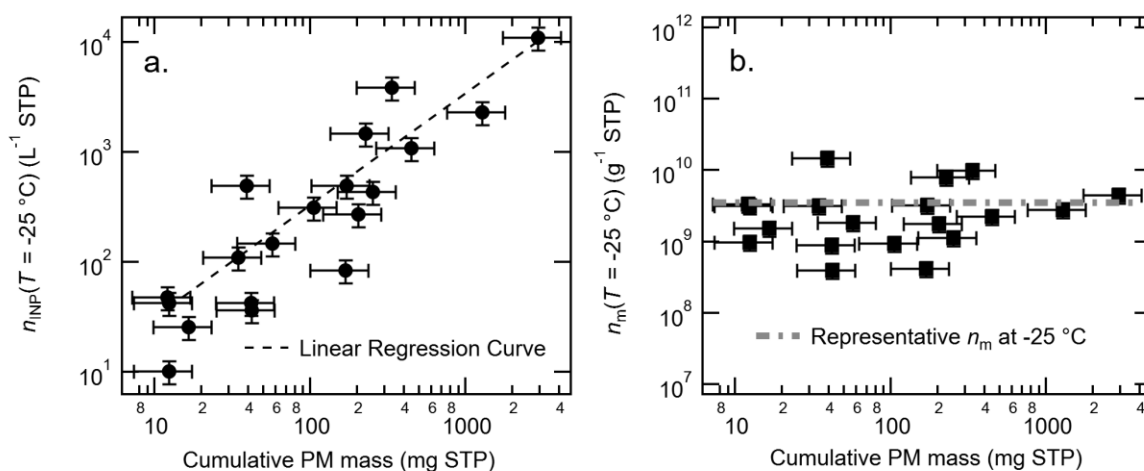


Figure 5. Correlation between cumulative PM mass vs. n_{INP} (a) and vs. n_m (b) at -25 °C; a linear regression curve in log scale ($n_{\text{INP}} = 3.51 \times \text{Cumulative PM Mass} - 2.41$; $r = 0.94$) is shown in (a), and the constant value of representative n_m at the given T ($3.55 \times 10^9 \text{ g}^{-1}$), which is a median n_m value of minimum – maximum, is shown in (b). Note the errors in cumulative PM mass are $\pm 40.4\%$ as discussed in Sect. 3.1. The uncertainty in n_{INP} and n_m is $\pm 23.5\%$.

RC: - P6L31-32: Again, how was the “INP scaled to mass” conversion achieved?

AR: Explained above with equations.

RC: - P6L33-34: The authors claimed that ambient meteorological conditions might not be determining factors for INP concentrations. What about the difference between different sampling sites? Is there any previous study drew the same conclusion?

AR: As all of our study sites are located in the close proximity in the same Texas Panhandle region, we do not expect inter-spatial variabilities of local meteorology. As discussed above, the ambient OLLF dust properties are not spatially uniform, and the emitting mechanism itself is not controllable as it highly depends on a unit of mobile livestock and perhaps feeding practices etc. The authors are not aware of any previous studies conducted in a similar set up. We have clarified that this observation applies for our study sites only.

“These results imply the following: (1) ambient meteorological conditions, summarized in **Table 1**, might not be determining factors for n_{INP} for our study sites;...”

RC: - P6L34: Please add “as” before “summarized”.

AR: Added. Thanks.

RC: - P6L38: “several hundred INPs L-1” seems very high. Is there any previous report of such superior IN activity of ambient dust? Besides, please specify the “notable correlation” with solid particle size distribution and IN activity data.

AR: This is a good question. Now, the revised manuscript discusses this point in **Sect. 3.6. Comparison to previous soil dust IN studies**. Please see our revised manuscript.

RC: - P6L49: What does “they” refer to? Who/what agree with who/what?

AR: We have rephrased the sentence to clarify this as; “As explained in **Sect. 2.4**, a series of diluted samples were examined in INSEKT. We made sure to assess overlapping T intervals in a series of measurements to see if $n_{s,\text{geo}}$ values from multiple measurements agree within CL95% and, if so, to merge the results together.”

RC: - P6L52-53: The conclusion drawn here is vague and speculative. Why does different sample source affect the IN ability?

AR: We meant to address that the different samples possess different physicochemical properties. To clarify this point, we have rephrased the whole sentence to;

“These results suggest that (1) there is a difference in the INP abundance between bulk (< 75 μm -sieved) and aerosolized/filtered-samples for TXD01 (\lesssim 6.5 μm ; **Table 3**) presumably due to different properties in particles of these two size subsets (6.5 – 75 μm and \lesssim 6.5 μm) and/or different amount of IN-active soil organic matter (Tobo et al, 2014), (2) different physicochemical properties found for our TXD05 samples may not impact their INP propensities, and (3) TXD05 might be more representative of atmospherically relevant dust (see **Table 2** and **SI Sect. S2**).”

RC: - P7L1-11: This paragraph should move to Sect. 2.

AR: Moved to **Sect. 2.4**.

RC: - P7L14-15: The difference between heated and unheated samples measured by INSEKT and DFPC exhibited different trend instead of “not apparent” as the authors stated, please explain.

AR: The referee is right. The authors decided to detail and clarify the comparison of the non-heat-treated sample to the heated-sample is discussed in **Sect. 3.6** and **SI Sect. S3**. Please see the revised manuscript.

RC: - P7L12-16: The comparison of the instruments is irrelevant to the purpose of this section and should go to SI. In addition, why are the results of WT-CRAFT not added to compare the three instruments simultaneously?

AR: WT-CRAFT was primarily used to analyze the field samples and not used for systematic measurements of TXD01 and TXD05. The instrument comparison discussion is now in SI Sect. S3.

RC: - P7L28-30: Again, the result and interpretation seem very speculative. Is there any physical/chemical/biological evidence to support the statement here?

AR: The authors agree. This speculative part and associated reference have been removed from the manuscript.

RC: - P7L32-33: How representative are the samples tested in this study to “natural supermicron-dominant INPs”? Again, very broad and general statement without support.

AR: This is a valid point. To reflect what is presented. The authors rephrased; natural supermicron-dominant INPs to: supermicron-dominant INPs from cattle feedyard, which can act as an important point source of agricultural INPs

RC: - P7L33-35: Did the authors compare the immersion parameterization proposed in this work with other widely used parameterization models? How consistent and different they are? Whether the new immersion parameterization is universal? Also: Fitting an immersion parameterization solely as a function of a single parameter, temperature, might be misleading.

AR: Offering a universal or world representative parameterization for agricultural INPs is not the scope of this work. As each OLLF represents a point source of fresh agricultural dust, we expect that it would have a different ice nucleation efficiency compared to aged/weathered dusts. Nonetheless, our new Fig. 8 shows a comparison of our $n_{s,geo}$ data with six relevant IN parameterizations of soil/desert dust. Our parameterization is comparable to these past agricultural soil IN parameterization but with some deviation. The $n_{s,geo}(T)$ parameterization scales to the total particle surface – so it can be parameterized as a function of T (Niemand et al., 2012), assuming that the time dependence can be neglected.

RC: The particle number and/or mass concentrations vary considerably around the world. Do the authors believe that the dataset of this parameterization is representative of the rest of the world?

AR: This is a good question – as responded in the last question, we did not intend to offer our parameterizations to represent agricultural INP across the world. Below we address in what sense(s), and to what extent, the dataset reported here can/cannot represent particulate-matter emissions from OLLFs the world over.

Qualitatively, it is reasonable to suppose that our TEOM data directly collected at the site represents mass concentrations of the fugitive aerosol particles because those aerosols derived from livestock manure and correspond to the dominant feedstuffs that the livestock have consumed. In this case, those dominant feedstuffs are steam-flaked corn (50-85% dry basis of the mixed diet), dry distiller’s by-products (0-30%), alfalfa hay (0-15%), and cereal silage (either corn or sorghum, 0-20%) (Asem-Hiablie et al., 2015), with various adjuncts, minerals, supplements, and fed pharmaceuticals present in relatively smaller amounts. To the extent that microbes of these aerosols are traceable to the microbial profile in the rumen, that behavior may be modulated by either the identity and relative amounts of the diet’s macro-components, or the nature and activity of any antimicrobials administered to the animals during their feeding period, or both. The species (i.e., *Bos taurus* vs. *Bos indicus*) and breed of the cattle may also influence aerosol characteristics through variations in optimal feed composition. Feeding practices for

beef cattle thus vary widely throughout the world. For these reasons, the authors can provide only a qualitative answer for this particular question.

Bush et al. (2014) presented, in a generalized form, the mathematical basis and workflow associated with estimating emission fluxes from an extensive area source. Taken together, that approach – to which many including Seibert (1998) have attached the name “inverse dispersion modeling,” or IDM – implicitly concedes several methodological criticisms that relegate the resulting emission-flux estimates to the status of *relative* quantities rather than *absolute* quantities. Those criticisms have been helpfully and succinctly summarized in Botlaguduru (2009). First, the emission-flux estimates are model-dependent; given a set of atmospheric conditions and measured mass concentrations as the model inputs, any two different dispersion models will yield two distinct estimates of the source strength. Second, because direct measurement of fugitive emissions fluxes from extensive area sources is not possible with current technology, it follows that an emission flux inferred via IDM with one dispersion model must not be used as the source-strength input to a different dispersion model operating in forward mode, for example, to predict downwind concentrations under other weather conditions. In other words, emission-flux estimates from IDM may be thought of as “correct” or “accurate” only in relative terms, and only with respect to the precise dispersion model used in the IDM workflow. Third, even the concept of an emission flux – an emission rate per unit area, having units of $M L^{-2} T^{-1}$ – is itself deeply problematic when used to quantify the source strength of an OLLF.

Importantly, a common computational approach in the refereed literature obscures the third deficiency. It has long been understood that hoof action on dry, uncompacted manure is the primary mechanism responsible for fugitive dust emissions from OLLFs. However, because dispersion models traditionally used with agricultural area sources are developed on the assumption of spatially uniform (or even piecewise spatially uniform) source strength, another attribute of OLLFs known as the stocking density – the average number of animals housed per unit area, with units of $\# (\text{head}) L^{-2}$ – was added as a scaling factor once the emissions flux had been estimated through IDM. The resulting, scaled source strength was given the name, “emission factor,” which according to the U. S. Environmental Protection Agency is “a representative value that attempts to relate the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant.” The implicit idea is that if the extensive magnitude of a polluting activity (PA) is known, and if the emission factor (EF) associated with that activity is known, then the emission rate or flux (Q) associated with that activity can be straightforwardly computed (EPA, 1995) as

$$Q = EF \times PA \qquad \text{Eqn. [A]}$$

In equation [A], which renders the EPA formula but omits the factor for control-method effectiveness, the units of both EF and PA are selected source-type by source-type (a) to represent the emission mechanism faithfully so that scaling by magnitude is methodologically valid and (b) to yield in their arithmetic product the desired units of the source strength, Q. In cattle feeding, given that fugitive emissions result from (and scale by, in approximate terms) animal activity rather than production area, the appropriate units for EF are $M (\text{hd}^{-1}) T^{-1}$, as in “lbs/hd/d.” Correspondingly, the polluting activity (PA) can be described by the stocking density, $\text{hd} L^{-2}$, as in “hd/ac.” The product of those two quantities yields a flux, $M L^{-2} T^{-1}$. However, in practice, the computation works the other way: Q is inferred from inverse dispersion modeling, with all of the methodological problems pertaining to it, PA is computed from an OLLF’s production records, and EF is computed from equation [A]. By now, it should be obvious that EF is itself subject to exactly the same criticisms as was Q; the scaling process through equation [A] was merely a fig leaf to present the emission factor, EF, as the fundamental quantity – having units appropriate to the emissions mechanism!– even though it was itself derived from the model-contingent quantity, Q.

The cumulative weight of those criticisms may leave the reader suspecting that the criticisms are overly tendentious and convenient. However, the history of the cattle feedlot emission factor in EPA's own guidance document confirms the criticisms' validity as a practical matter. That guidance document, cited above as "AP-42," includes a set of quality ratings for the emission factors contained in it as guidance for state air pollution regulatory authorities. For many years (EPA, 1985), the AP-42 emission factor for cattle feedlots, a subtype of OLLFs, was rated "Class E (poor)," which denoted that the emission factor "is developed from C- and D-rated test data, and there may be reason to suspect that the facilities tested do not represent a random sample of the industry...there also may be evidence of variability within the source category population." Quite so, as cited research abundantly confirms. Still, EPA retained its AP-42 guidance for particulate-matter emissions from cattle feedlots until 1995, after which even that Class E emission factor has been omitted.

RC: - P7L44-45: Previous studies on bio-aerosol INP activity reported the importance OF ice active protein instead of DNA. Why didn't the authors conduct protein analysis?

AR: The ice nucleation protein is a minor protein of the outer membrane of bacteria with ice nucleation activity, about 1% of outer membrane proteins only (Morris et al., 2004) and therefore hard to use as a target in bioaerosol studies. DNA-based detection offers greater sensitivity because, theoretically, if the DNA of even one cell in a sample is successfully extracted, it will be amplified and detected in a metagenomics study. Moreover, the research team which produced this manuscript does not possess the instrumentation for protein detection.

RC: - P7L59: The data is not consistent with Table 7.

AR: Corrected. Thanks for catching this.

RC: - P7L6-9: Why do the authors refer to previous study instead of the single particle chemical analysis in this study to infer particle hygroscopicity?

AR: That is because we previously found there is an inherent relationship between the composition and the hygroscopicity of OLLF particles. We clarified this sentence as, "Higher aspect ratios in residuals compared to aerosol particles were found for both TXD01 and TXD05 samples. This difference indicates a relative increase in non-spherical particles, that have a higher aspect ratio, in residuals. In short, Hiranuma et al. (2008) found that quasi-spherical OLLF particles were predominantly salt-rich hygroscopic particles, whereas non-spherical amorphous particles were found to be organic-dominant with negligible hygroscopicity. Thus, our results suggest the inclusion of non-hygroscopic particles as ice residuals."

RC: - P7L17-18: Data presented in Table 8 does not support this statement. Why did the authors decide to omit the influence of carbonaceous content on particle IN activity?

AR: We did not omit the influence of carbonaceous content. EDX provides only elemental composition (no molecular-level info), and almost all of our particles contained organics, which is consistent with our previous study with Raman (H11). We believe that we addressed in text corresponds to what we show in Table 8 with up and down arrow marks. In addition, the authors decided to address the importance of future work regarding soil dust-derived organic INPs in Sect. 4 as, "...Based on findings from this study, ICR analysis revealed a relative increase in organic inclusion (and decrease in salt inclusion) in residuals, highlighting the importance of organic material in OLLF-derived INPs for atmospheric immersion. Even after dry heating treatment, the increase in organic fraction was found in the ICR of our OLLF samples. Therefore, the investigation of heat-insensitive organics is key to further understand the properties of soil dust INPs, and further research should focus on understanding how organic composition influences IN. Our previous work using Raman micro-spectroscopy revealed that ambient aerosol particles sampled at OLLFs are internally mixed with brown or black carbon, hydrophobic humic acid, water soluble organics,

less soluble fatty acids, and carbonaceous materials mixed with salts and minerals. But, our current knowledge regarding IN-active organics is still limited.”

RC: - P7L38: Why did the authors use different units for PM mass loading here?

AR: We would like to offer both units used in Eqn (1), g L^{-1} , and what TEOM offers as a preset unit, $\mu\text{g m}^{-3}$.

RC: - P7L47: What is the significance of average estimated INP concentrations between different years?

AR: This is an interesting question. We now provide the visualized inter-annual variability of average estimated INP concentrations in Fig. 9.

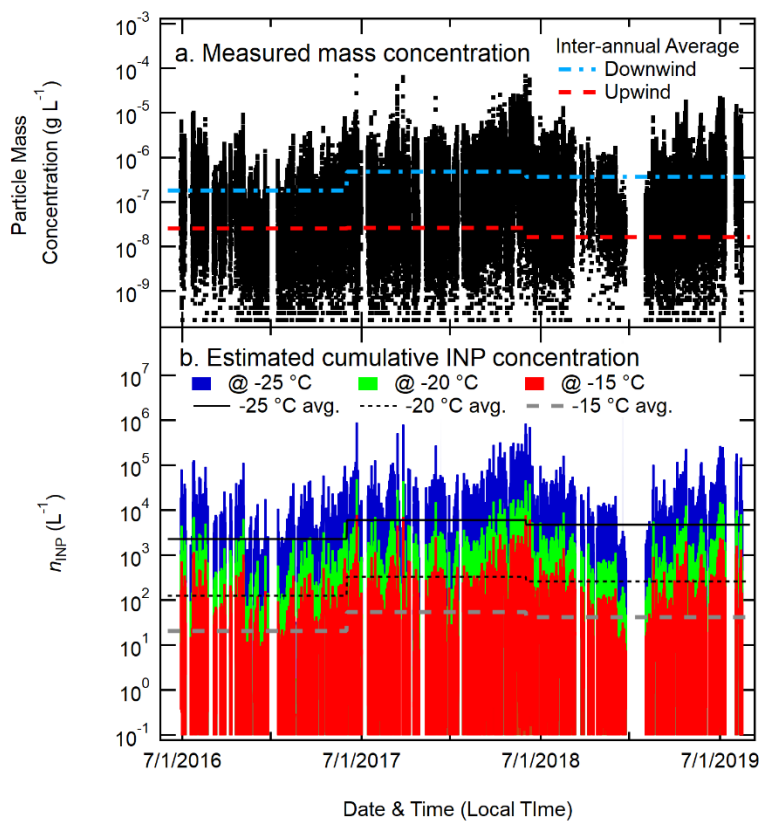


Figure 9. OLLF INP concentrations. Time-series plot of TEOM mass concentration measured at the downwind side of OLLF-1 (a) and cumulative n_{INP} estimated at T s of $-15\text{ }^{\circ}\text{C}$, $-20\text{ }^{\circ}\text{C}$, and $-25\text{ }^{\circ}\text{C}$ (b). In Panel a, inter-annual average mass concentrations of aerosol particles from OLLF (blue dashed line) and upwind (red dashed line) are shown (numbers adapted from Table 8). In Panel b, likewise, inter-annual average n_{INP} estimated at -15 , -20 , and $-25\text{ }^{\circ}\text{C}$ (reported in Table 8) are also shown. Meteorological summer in Texas is used for the beginning and ending time stamps of each year.

Furthermore, the authors decided to discuss the inter-seasonal variations in estimated PM and INP concentrations using Table 8 in Sect. 3.5.

Table 8. Inter-annual and seasonal PM_{10} mass concentrations from OLLF-1 as well as estimated n_{INP} .

	PM_{10} Mass Concentration (g L^{-1})		Estimated $n_{\text{INP}}(T)$ (L^{-1})		
	*OLLF	Upwind	$T = -15\text{ }^{\circ}\text{C}$	$T = -20\text{ }^{\circ}\text{C}$	$T = -25\text{ }^{\circ}\text{C}$
2016 – 2017	1.8E-07	2.6E-08	20.7	127.5	2323.4

Summer	3.7E-07	5.2E-08	42.3	260.5	4747.7
Fall	1.6E-07	2.8E-08	18.1	111.7	2036.3
Winter	6.3E-08	1.5E-08	7.2	44.2	806.2
Spring	1.6E-07	2.1E-08	17.7	108.9	1985.5
2017 – 2018	4.8E-07	2.6E-08	54.6	336.4	6133.0
Summer	3.0E-07	2.3E-08	33.8	208.5	3801.1
Fall	3.1E-07	1.9E-08	35.4	218.2	3978.3
Winter	2.5E-07	1.3E-08	27.9	171.7	3129.6
Spring	9.2E-07	4.6E-08	104.1	641.3	11690.9
2018 – 2019	3.7E-07	1.7E-08	42.3	260.7	4752.5
Summer	4.9E-07	2.6E-08	55.6	342.3	6240.6
Fall	2.4E-07	7.9E-09	26.8	165.3	3013.0
Winter	1.5E-07	1.3E-08	17.0	104.8	1910.2
Spring	2.8E-07	1.6E-08	31.8	195.8	3570.0

*Upwind concentration is subtracted.

The associated text in Sect. 3.5 has been rephrased as, “In general, PM₁₀ mass concentrations from OLLF-1 (average ± standard errors) were high in meteorological summers ($3.9 \times 10^{-7} \pm 5.6 \times 10^{-8} \text{ g L}^{-1}$) and springs ($4.5 \times 10^{-7} \pm 2.4 \times 10^{-7} \text{ g L}^{-1}$) as compared to fall ($2.4 \times 10^{-7} \pm 4.4 \times 10^{-8} \text{ g L}^{-1}$) and winter ($1.5 \times 10^{-7} \pm 5.3 \times 10^{-8} \text{ g L}^{-1}$). A similar trend was found for the upwind PM₁₀ mass concentration: summer ($3.4 \times 10^{-8} \pm 9.0 \times 10^{-9} \text{ g L}^{-1}$) ≥ spring ($2.8 \times 10^{-8} \pm 9.3 \times 10^{-9} \text{ g L}^{-1}$) > fall ($1.8 \times 10^{-8} \pm 5.7 \times 10^{-9} \text{ g L}^{-1}$) ≥ winter ($1.4 \times 10^{-8} \pm 7.1 \times 10^{-10} \text{ g L}^{-1}$). But, the measured values at the upwind location are consistently an order magnitude lower than that from the downwind location.

On average, the estimated mean n_{INP} values at -15, -20, and -25 °C in 2016 – 2019 were estimated as 46.8 (±25.3 seasonal standard deviation; same hereafter), 288.1 (± 156.1), and 5,250.9 (± 2,845.6) L⁻¹, respectively. In addition, the median n_{INP} at -15, -20, and -25 °C in 2016 – 2019 were estimated as 14.7 (± 9.2), 90.9 (± 56.4), and 1,656.3 (± 1,028.1) L⁻¹, respectively. As our n_{INP} is linearly scaled to mass concentration (Eqn. 1), estimated n_{INP} showed a similar seasonal variability as seen in mass concentration. For instance, at -20 °C, the cumulative n_{INP} averages for each meteorological season over three 2016 – 2019 were estimated as follows: spring ($315.4 \pm 164.9 \text{ L}^{-1}$) ≥ summer ($270.4 \pm 39.0 \text{ L}^{-1}$) > fall ($165.1 \pm 30.8 \text{ L}^{-1}$) ≥ winter ($106.9 \pm 36.8 \text{ L}^{-1}$). The observed high n_{INP} values were expected for such a high PM₁₀ mass concentrations emitted from the cattle feedyard, which represent an important point source of agricultural aerosol particle emission. However, we reemphasize that the IN efficiency of OLLF aerosol particles is somehow similar to other agricultural aerosol particles found in previous studies as discussed in Sect. 3.2 (Fig. 8).“

RC: - P7L490-50: Again, is there any reason for such high INP activity? Is there any previous paper on such efficient INP?

AR: Addressed above.

RC: - P7L52-53: Reads like introduction.

AR: This sentence is removed.

RC: - P8L4-35: Please rephrase and improve the writing quality.

AR: Done.

RC: - Please replace “lpm” with “LPM” in the paper and check unit usage throughout.

AR: Replaced.

RC: - Please check the space between the number and °C for consistency.

AR: Done.

RC: Fig. 1:

Panel (c): What does the green shading in panel (c) stand for?

AR: RH uncertainty – “RHs were determined with an accuracy of $\pm 5\%$, represented as green shaded area in (c), using the mean gas T and the mean water vapor concentration.”

Why did the RH drop below water saturation in column (ii-iv), (vi), (viii), and (ix) during immersion freezing studies?

AR: This is a valid question. At these T_s , ice crystals grow rather fast at an expense of available water vapor in the AIDA chamber, which causes a drop in RH. Nevertheless, droplets should be fully activated within first 100 seconds of each expansion until the peak RH value is reached (Fig. 7). Moreover, as seen in panels (d), the number concentration of particles with $>20 \mu\text{m } D_{ve}$ is not increasing after the RH peak, and all predominant ice formation occurs at or before the RH peak through immersion freezing. Lastly, as seen in Fig. 8, we made sure to only report our IN efficiency at T_s higher than $\sim -30^\circ\text{C}$, corresponding to water saturated condition in the AIDA vessel. This point is now clarified in Sect. 3.2 (please refer to the track changes version manuscript).

Panel (d): The total particle concentration line is hard to notice.

AR: We have modified all d panels with the right y-axis assigned for total particle concentration.

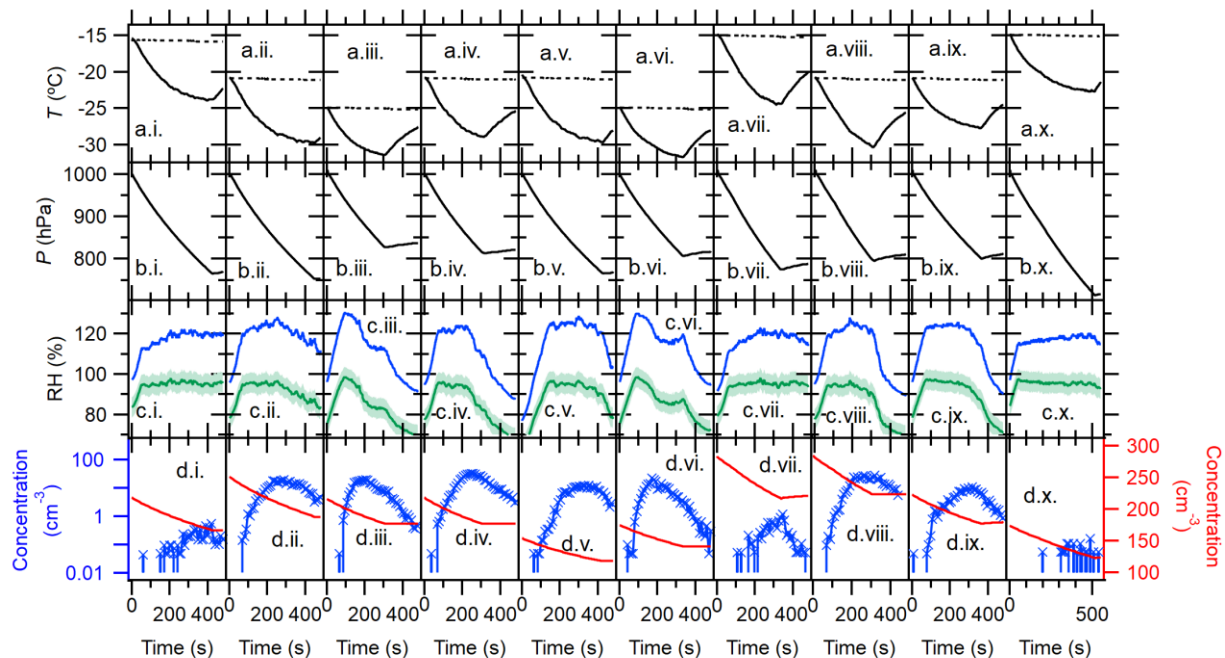


Figure 7. Temporal profiles of the AIDA immersion freezing experiment [TXDUST01_07 (i), _08 (ii), _30 (iii), _12 (iv), _13 (v), _32 (vi), _3 (vii), _4 (viii), _16 (ix), _17 (x)]. Arrays of alphabetical panels represent the chamber gas T (solid line) and the chamber wall T (dashed line) (a), P in the AIDA chamber vessel (b), RH with respect to water (green line) and ice (blue line) (c), and aerosol particle concentration initially measured by the CPC (red solid line) as well as number concentration of $> 20 \mu\text{m } D_{ve}$ AIDA particles measured by a welas optical particle counter (blue line) (d). Horizontal numerical panels represent different sample types and AIDA experiments, including TXD01 (i) – (iii), TXD05 (iv) – (vi), TXD01H (vii) –

viii), and TXD05H (ix – x). RHs were determined with an accuracy of $\pm 5\%$, represented as green shaded area in (c), using the mean gas T and the mean water vapor concentration.

This figure add little to the paper, should it go to SI?

AR: It is a valid suggestion. While we put minimum text associated with this figure, the authors believe that this figure represents important result of AIDA experiments. We would like to keep this in the main manuscript.

RC: Fig. 2: The font size is too small and unclear.

AR: The former Fig. 2 is now moved to SI Sect. S1 (Fig. S1). The authors believe that all texts are visible in Fig. S1.

P14L13-15: Please rephrase.

AR: Rephrased.

P14L13: What is “CF-to-IF” ratio?

AR: counterflow to input flow ratio

This figure add little to the paper, should it go to SI?

AR: The authors concur. Now, it is moved to SI Sect. 1.

RC: Fig. 3: The font size is too small and unclear. Labels of panels a and b are missing.

AR: As discussed above, the authors decided to further analyze our data for spatial variability (Fig. 3) and seasonal variability (Figs. 4-6) to explain the observed data variation and completely reformulated Sect. 3.1. Please see the revised figures in the track changes manuscript.

RC: What does n_s in the figure refer to? The authors mentioned $n_{s,geo}$ in P6L11, is there any relevance between the text and this figure? Why is the caption inconsistent with the figure? Is there any difference among $n_{s,geo}$ (STP), $n_{s,geo}$, n_s (STP), and (STP)?

AR: We now strictly limited our terminologies to $n_{s,geo}$ and $n_{s,geo}(T)$, which is used for the $n_{s,geo}$ as a function of temperature, throughout the manuscript. They are all computed for STP.

RC: Fig. 4: Please make it explicit that this figure is for -25°C .

AR: Good point. Done.

RC: What caused such large error of cumulative PM mass?

AR: This is a valid question. The errors in cumulative PM mass are on average 40.4%, derived from calibration of two DustTrak instruments against TEOM in a side-by-side position. This error mainly stems from variable emission flux at OLLF rather than a systematic bias of DustTrak ($<10\%$; Wallace et al., 2011). Briefly, the aerosol particle emission potential at OLLF is associated with highly pulverized, nearly single-grained surface material that develops on OLLF pen surfaces as manure accumulates, dries, and is crushed by animal hoof action, as contrasted with the small to medium-sized “clods” from which the single-grained material ultimately develops. Bush *et al.* (2014) provide a more extensive account of fugitive-dust emission dynamics and their interaction with boundary-layer stability to create transient peaks in ground-level mass concentrations of fugitive dust. Again, as addressed above, it is important to note that the emission “flux” at OLLF is not spatially uniform (not even in a *piecewise* sense), and the emitting mechanism itself is not a property of a source area *per se* but of a unit of mobile livestock. Granting the

primacy of hoarfrost action as the decisive emissions mechanism as described above and in Bush *et al.* (2014), a more accurate representation of the source strength of an OLLF would be e. g. an agent-based model in which the source is the time-varying aggregation of many mobile point sources, as described by Auvermann (2003). We also shorten the new Fig. 5 caption to “Note the errors in cumulative PM mass are $\pm 40.4\%$ as discussed in Sect. 3.1.”

RC: “The uncertainty in nINP and nm is $\pm 23.5\%$ ”. However, the error bars of nINP in Fig. 4(a) seems not equal to this value. Also: the correlation coefficient and/or other parameters should be given in Fig. 4.

AR: Double checked – they are good.

RC: Fig. 5: The color of error bars is the same with the color of heated samples. Please make this figure clearer for readers.

AR: Modified.

RC: Why did the authors use different scales in the same figure?

AR: Now, we incorporated all in the same scale.

RC: The meaning of ns,geo is different from the definition in P6L11. Please clarify if these are different quantities, and explain why the terms and symbols are so confusing in the paper.

AR: Clarified in the manuscript, and it is now consistent.

RC: Please check the error bars in panel c. How come that the upper error bars are longer than the lower bars in a log-scale plot?

AR: Checked.

RC: Please check the labels of the figures for consistency.

AR: Checked.

RC: In Fig. 5 (a.ii), there is a significant difference between the filter and bulk samples at temperatures above $-20\text{ }^{\circ}\text{C}$, but no such difference in Figure 5 (b.ii). What causes this difference?

AR: Explained in Sect. 3.2.

RC: Fig. 6: Is it worth wasting a figure and report just one set of DFPC data rather than include the data in Fig. 5?

AR: The referee has the point. We now merged a subset of DFPC data in Fig. 8.

RC: Table 2: Why did the author report “relative standard deviation of $\pm 3\%$ ” instead of one standard deviation from the mean value?

AR: No special intension, we thought % would be more straightforward to the reader. We now report standard deviation instead of relative one.

RC: How does heating affect ATD sample density?

AR: The authors do not discuss anything about ATD here.

RC: Why did heating lead to increased density for TXD1?

AR: For the given uncertainty, TXD1 and TXD1H show similar ice nucleation efficiencies. So it is not conclusive

RC: Why didn't the author measure the density of a specific sample before and after heating?

AR: The authors measured the density of all bulk samples before and after heating as shown in Table 2 and discussed in Sect. 2.2. We wanted to make sure that physical properties, like the density, are not impacted by heating.

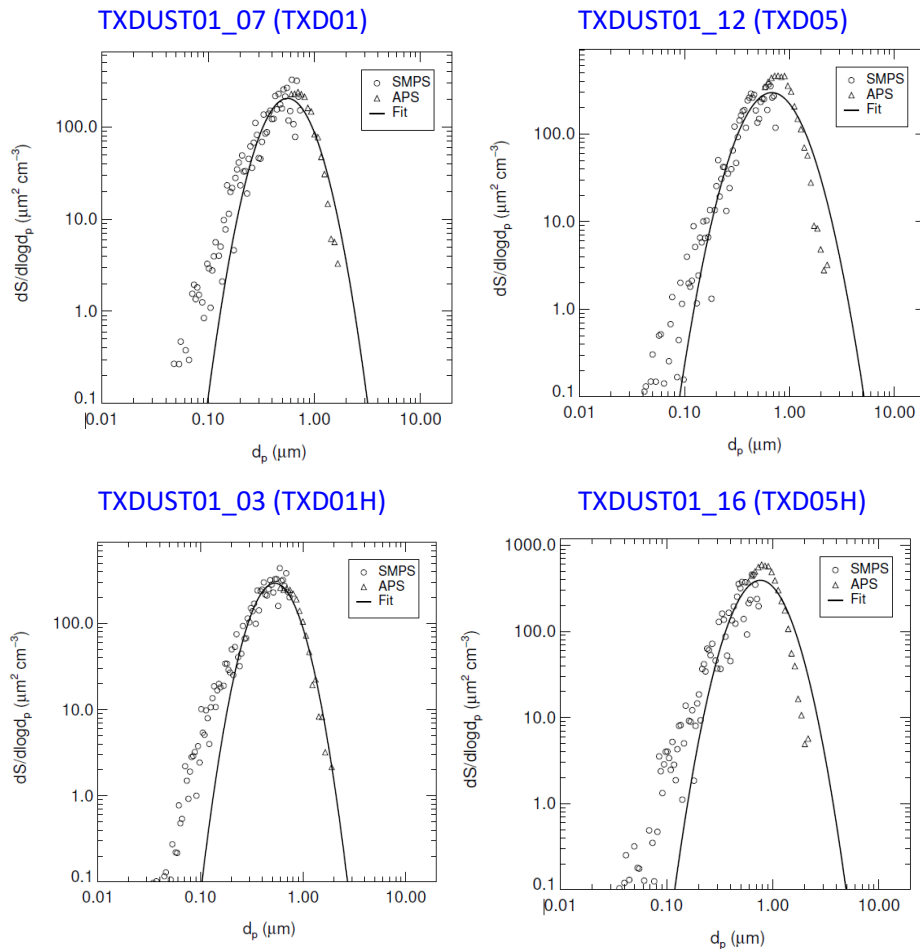
RC: Table 3: What does the number in Experiment ID mean? It's very hard to understand together with Fig. 1.

AR: Unique code used in AIDA. In Sect. 3.2, now we clarify this code as, "All lab data associated with this study were archived according to the AIDA experiment number (i.e., TXDUST01_number), and we share these IDs for other associated measurements (e.g., INSEKT)."

RC: Why is the size distribution so wide? What does the size distribution look like?

AR: We did not select the size. It is polydisperse in nature.

We show the particle surface area distribution of TXDUST01_07, _12, _3, and _16 as for snapshot examples of each sample type (TXD01, TXD05, TXD01H, and TXD05H, respectively) here.



Why didn't the author report particle number size distribution? What quantities do the letters refer to?

AR: We use surface (S) to estimate ice nucleation efficiencies, so we report only surface-based distribution.

$N_{\text{total},0}$ = total number concentration of particles at the initial stage ($t = 0$) prior to expansion.

$S_{\text{total},0}$ = total surface concentration of particles at the initial stage ($t = 0$) prior to expansion.

$M_{\text{total},0}$ = total mass concentration of particles at the initial stage ($t = 0$) prior to expansion.

D_{ve} = volume equivalent diameter.

RC: Table 4: Why do the authors put a table here with little description and discussion in text?

AR: Now, the discussion is extended based on the referee's suggestion (i.e., comparison to previous studies), and we would like to keep this table in this main manuscript.

RC: Table 7: Why were the residual projected sizes consistently larger than those of aerosols except for TXD05H? What do the authors think happened here?

AR: This was expected as the larger the particles, the more surface is available for ice nucleation to take place. The reason for the TXD05H exception is not known. Fig. S2 shows >10% reduction of K-rich particles (only 2% for TXD01H) so, loss of condensation active INP may play a role. But as this is not conclusive, we will limit the discussion regarding TXD05H.

Other revisions:

West Texas → the Texas Panhandle

The authors recently learned that ACP does not accept “West Texas” as a proper noun. As the place name needs to have clearly defined boundaries and be internationally known, we decided to adapt “the Texas Panhandle” and replaced it with all West Texas in this paper. At its first appearance in the manuscript, we define it as “the Texas Panhandle (northern most counties of Texas; also known as West Texas)”.

Dr. Larissa Lacher has been added as a coauthor because she has visited the Texas Panhandle to in part support the field sampling activities at OLLFs and was involved in analyzing some INSEKT samples, which led to improve SI Sect. S3.

We have a new acknowledgements section and separated financial support statements.

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