

Review of “Feedlot is a unique and constant source of atmospheric ice-nucleating particles” by Hiranuma et al.

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.

Major comments:

1. Introduction: The logic of this part needs to be thoroughly improved. A much more detailed review of relevant previous work should be added. 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? How representative (e.g. size distribution, chemical composition, etc.) are the surface samples collected in this study compared to actual ambient aerosols? How representative are the sampling sites for the whole Texas? The title made a very broad and general statement without any solid support.
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.
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.

Specific comments:

- P1L18: Please capitalize each word.
- P1L21: Please add “the” before U.S. and check the usage of “the” throughout.
- P1L28: “thereby finding...” is not clear to me.
- P1L30: Please replace “improved” with “improving”.
- P1L32: Please rephrase to “...explore the relationship between INP and supercooled cloud properties...”.

- P1L38: 25% of total what?
- P1L46: Please rephrase.
- P1L48: “suggesting their regional scale impact” is not clear to me.
- P1L50: Please verify the usage of “impact in”.
- 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.
- P1L54: Please add the classical textbook of Pruppacher and Klett (2010). Please also spell out “INP”.
- P1L56: Please rephrase “focused in this study”.
- P2L14: Please spell out “Ts” or specify the meaning of the subscript.
- P2L15: Please spell out “ICR”.
- P2L39: Please add “as” before addressed.
- P2L43: Please spell out “AIDA”.
- P2L47: Please spell out “TXD”.
- P2L58: What instrument did the author use to measure BET SSA?
- 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?
- 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?
- P3L15: Please see the comment above.
- P3L17: What is the purpose of filter sampling? Can the authors provide a scheme of the experimental setup in this study?
- P3L24: Please add reference after “IS-PCVI”.
- P3L28: Should be μm here.
- P3L31: Does the authors mean “individual ice crystal” here? If so, maybe it’s better to delete “of individual particles”.
- P3L41: Please add more AIDA references apart from the review paper.
- 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?
- P3L45: What technique did the authors use to aerosolize particles?
- P4L2: Did the authors intend to say “array”?
- P4L7: Please spell out “HPLC”.
- P4L8-9: How was the detection limit determined?
- P4L11-12: Can the authors provide an example for such processing in SI?
- P4L25-26: If it’s “a series of diluted suspensions”, should it be “ $\times 15$ to $\times 225$ ” here? Besides,

please replace “x” with “×” throughout the paper.

- P4L38: Remove the “of” after “100 L”.
- P4L40: “was characterized”.
- P4L42: Please specify what type of “diameter”.
- Sect. 2.6: Please simplify the description of previous work.
- P5L31: What is the typical size range of droplets and ice crystals in this study?
- P5L36: Should be μm . Please check the units throughout.
- P5L37: During TXDUST01 what?
- P5L45: Please change the parenthesis to “(...Model 1400a; Patashnick...)”.
- P5L58: Please enclose “ $<100 \mu\text{g m}^{-3}$ ” into parenthesis.
- P6L3-9: What’s the point of repeating previous results in this paper?
- 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.
- P6L22: How were these quantities derived? Can the authors please provide the derivation or conversion process in SI apart from the references here?
- P6L23: Please verify the usage of “Regardless”.
- P6L25: How did the author decide whether the data was “uncertain systematically erroneous data”?
- 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.
- 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.
- P6L31-32: Again, how was the “INP scaled to mass” conversion achieved?
- 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?
- P6L34: Please add “as” before “summarized”.
- 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.
- P6L49: What does “they” refer to? Who/what agree with who/what?
- P6L52-53: The conclusion drawn here is vague and speculative. Why does different sample source affect the IN ability?
- P7L1-11: This paragraph should move to Sect. 2.

- 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.
- 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?
- P7L28-30: Again, the result and interpretation seem very speculative. Is there any physical/chemical/biological evidence to support the statement here?
- P7L32-33: How representative are the samples tested in this study to “natural supermicron-dominant INPs”? Again, very broad and general statement without support.
- 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. 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?
- 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?
- P7L59: The data is not consistent with Table 7.
- P7L6-9: Why do the authors refer to previous study instead of the single particle chemical analysis in this study to infer particle hygroscopicity?
- 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?
- P7L38: Why did the authors use different units for PM mass loading here?
- P7L47: What is the significance of average estimated INP concentrations between different years?
- P7L490-50: Again, is there any reason for such high INP activity? Is there any previous paper on such efficient INP?
- P7L52-53: Reads like introduction.
- P8L4-35: Please rephrase and improve the writing quality.
- Please replace “lpm” with “LPM” in the paper and check unit usage throughout.
- Please check the space between the number and °C for consistency.

Fig. 1:

Panel (c):

- What does the green shading in panel (c) stand for?
- Why did the RH drop below water saturation in column (ii-iv), (vi), (viii) and (ix) during **immersion freezing** studies?

Panel (d):

- The total particle concentration line is hard to notice.

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

Fig. 2:

- The font size is too small and unclear.
- P14L13-15: Please rephrase.
- P14L13: What is “CF-to-IF” ratio?

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

Fig. 3:

- The font size is too small and unclear.
- Labels of panels a and b are missing.
- 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)?

Fig. 4:

- Please make it explicit that this figure is for -25°C .
- What caused such large error of cumulative PM mass?
- “The uncertainty in n_{INP} and nm is $\pm 23.5\%$ ”. However, the error bars of n_{INP} in Fig. 4(a) seems not equal to this value. Also: the correlation coefficient and/or other parameters should be given in Fig. 4.

Fig. 5:

- The color of error bars is the same with the color of heated samples. Please make this figure clearer for readers.
- Why did the authors use different scales in the same figure?
- The meaning of $n_{s,geo}$ 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.
- 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?
- Please check the labels of the figures for consistency.
- In Fig. 5 (a.ii), there is a significant difference between the filter and bulk samples at temperatures above -20°C , but no such difference in Figure 5 (b.ii). What causes this difference?

Fig. 6:

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

Table 2:

- Why did the author report “relative standard deviation of $\pm 3\%$ ” instead of one standard

deviation from the mean value?

- How does heating affect ATD sample density? Why did heating lead to increased density for TXD1? Why didn't the author measure the density of a specific sample before and after heating?

Table 3:

- What does the number in Experiment ID mean? It's very hard to understand together with Fig. 1.
- Why is the size distribution so wide? What does the size distribution look like?
- Why didn't the author report particle number size distribution? What quantities do the letters refer to?

Table 4:

- Why do the authors put a table here with little description and discussion in text?

Table 7:

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

Reference

Pruppacher, H. R., and Klett, J. D.: *Microphysics of Clouds and Precipitation*, 2 ed., Atmospheric and Oceanographic Sciences Library, 18, Springer Netherlands, XXII, 954 pp., 2010.