TNA User Report

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Project title	AIDA (Aerosol Interaction and Dynamics in the Atmosphere) investigation on the ice nucleation activity of dust particles emitted from cattle feeding operations in the Texas Panhandle
Name of the accessed chamber	AIDA
Number of users in the project	5
Project objectives (max 100 words)	The overall objective of the study is **characterizing physical, chemical and biological properties of ice-nucleating particles (INPs) from a cattle feedlot in Texas, USA**, and their relation to **ice nucleation processes, especially immersion freezing,** in simulated cloud systems using the AIDA chamber. New data on the ice nucleation (IN) properties of agricultural dust at heterogeneous freezing temperatures will be generated, including **assessment of heat influence on abundance and composition of feedlot INPs**.
Description of work (max 100 words)	This study combines an investigation of the ice-nucleating propensity of INPs by AIDA cloud chamber experiments, ice crystal residual (ICR) analyses and other modern suite of online and offline aerosol characterization instruments for the first time for the feedlot sample. Our INP and ICR measurements will be useful to generate new IN parameterizations that would help predict primary ice crystal concentrations representative for the particle-laden agricultural source. Sample analyses are just beginning.

Principal Investigator's and group's information	
First name	Naruki
Family name	Hiranuma
Nationality	South Korean/Japanese
Activity domain[1]	Earth Sciences & Environment
Home institution	West Texas A&M University
Institution legal status[2]	UNI
Email	nhiranuma@wtamu.edu
Gender	Male
User status[3]	ACA
New user	Yes

User 1 Information[4]	
First name	Brent
Family name	Auvermann
Nationality	American
Activity domain	Earth Sciences & Environment
Home institution	Texas A&M AgriLife Research at Amarillo
Institution legal status	RES
Email	BAuverma@ag.tamu.edu
Gender	Male
User status	RES/ENG/ACA
New user	Yes

User 2 Information	
First name	Kimberly
Family name	Cory
Nationality	American
Activity domain	Earth Sciences & Environment
Home institution	West Texas A&M University
Institution legal status	UNI
Email	kmcory1@buffs.wtamu.edu
Gender	Female
User status	PGA
New user	Yes

[1] Physics; Chemistry; Earth Sciences & Environment; Engineering & Technology; Mathematics; Information & Communication Technologies; Material Sciences; Energy; Social sciences; Humanities.
[2] UNI= University and Other Higher Education Organisation;
RES= Public Research Organisation (including international research organisations and private research organisations controlled by public authority);
SME= Small and Medium Enterprise;
PRV= Other Industrial and/or Profit Private Organisation;
OTH= Other type of organization.
[3] UND= Undergraduate; PGR= Post graduate; PDOC= Post-doctoral researcher; RES= Researcher ENG= Engineer; ACA= Academic; TEC= Technician.
[4] Reproduce the table for each user who accessed the infrastructure

User 3 Information	
First name	Dimitri
Family name	Georgakopoulos
Nationality	Greek
Activity domain	Earth Sciences & Environment
Home institution	Agricultural University of Athens, Greece
Institution legal status	UNI
Email	dgeorga@aua.gr
Gender	Male
User status	ACA
New user	Yes

User 4 Information	
First name	Franco
Family name	Belosi
Nationality	Italian
Activity domain	Earth Sciences & Environment
Home institution	CNR-ISAC Istituto di Scienze dell'Atmosfera e del Clima
Institution legal status	RES
Email	f.belosi@isac.cnr.it
Gender	Male
User status	RES
New user	Yes

Trans-National Access (TNA) Scientific Report

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Name of the PI: Naruki Hiranuma
Chamber name and location: AIDA, Karlsruhe Institute of Technology, Karlsruhe, Germany
Campaign name and period: TXDUST01, 08-10-2018 to 26-10-2018

1. Introduction and motivation

Overview: The Texas Panhandle is a major contributor to the U.S. cattle production, accounting for 30% of total cattle population in Texas (>11 million head, *USDA*, 2012). Agricultural dust particles from animal feeding operations have been long known to affect regional air quality in the Texas Panhandle (*Von Essen and Auvermann*, 2005). In particular, open-air feedlots (OAFs) within 100 miles of West Texas A&M University represent a significant source of dust particles. The emission flux of PM_{10} (i.e., particulate matter smaller than 10 μm in diameter) from OAFs in this region exceeds 4.5 μg m^{-2} s^{-1} up to 23.45 μg m^{-2} s^{-1} depending on a stacking density (*Bush et al.*, 2014). However, their impact in cloud microphysics, especially ice nucleation (IN), is overlooked and poorly constrained although this region in the U.S. Midwest is dominated by Deep Convective Clouds where ice-nucleating particles (INPs) play a crucial role in precipitation and thunderstorm processes (*Li et al.*, 2017 ; *Rosenfeld et al.*, 2008). To fill this gap, **we comprehensively looked into immersion mode freezing abilities (*Vali et al.*, 2015) and other important properties, including physical, chemical and biological ones, of feedlot surface materials sampled at commercial and research OAFs in Texas**.

Motivation: Our preliminary field measurements in the Texas Panhandle show broad size distribution of ambient particles, and the INP concentration of approximately 10 L^{-1} at -10 °C, suggesting an inclusion of biogenic components (*Whiteside et al.*, 2018). Interestingly, **there is no notable correlation between INP and ambient aerosol concentrations** from our field study. This motivates the need for further characterization of our OAF samples in a controlled-lab setting and systematically identify what particulate features trigger their IN.

State-of-the-Art Properties of OAF Dust: To date, our previous work using Raman micro-spectroscopy revealed that ambient dust from OAFs is composed of brown or black carbon, hydrophobic humic acid, water soluble organics, less soluble fatty acids and those carbonaceous materials mixed with salts (*Hiranuma et al.*, 2011). But, **our knowledge regarding the abundance and composition of biological compartments of OAF dust is still scarce**. On average, a beef animal produces 82 lb per day (wet or as-is basis) of manure that is a complex microbial habitat, containing bacteria and other microorganisms, and is the predominant source of OAF dust when dried (*Mukhtar*, 2007; *Von Essen and Auvermann*, 2005). For instance, the cattle manure hosts a wide variety of bovine rumen bacteria (i.e., *Prevotellaceae*, *Clostridiales*, lipoprotein components of certain bacterial cell walls) as well as non-bacterial fauna of the rumen, such as fungal spores, lichens, fungi, plantae, protista, protozoa, chromalveolata and archaea (*Nagaraja*., 2016). **In this study, we targeted deductively determining the key cattle bovine microorganisms that, when aerosolized, act as INPs in the atmosphere.**

2. Scientific objectives

Our specific objective is to answer the following five research questions. Our hypotheses are described in the **Supplement Sect. S.1**.

a. What particulate features of OAF dust trigger IN in high-temperatures (Ts)?
b. What are the INP fractions of segregated submicrometer particles (<1 μm)?
c. How do ambient INP results compare to samples of feedlot surface materials?

d. How does cattle diet influence INP abundance in samples of feedlot surface materials?
e. What are the contributions of OAF particle properties to its INP propensity?

3. Reason for choosing the simulation chamber/calibration facility

We chose the AIDA as our study platform because this chamber is the world's foremost chamber for studying ice clouds in a controlled setting with respect to both *T* and saturation, at Karlsruhe Institute of Technology (KIT) (e.g., *Möhler et al*., 2003). This chamber generates artificial clouds and activates particles such that it simulates atmospheric cloud parcel via expansion cooling in a 84 m^3 vessel where the air volume adjacent to the chamber wall is too small compared to the rest in the vessel to neglect the wall effect (e.g., particle wall deposition). Using this instrument is critical to minimize error sources in INP observational data. The AIDA has been successfully applied for the analysis of both ambient and laboratory-generated INPs and has facilitated characterization of hundreds of INP species (*Hoose and Möhler*, 2012). Note that the AIDA results provided a validation of the other INP spectrometers employed in this study (i.e., INSEKT, DFPC, WT-CRAFT).

Another reason for choosing the AIDA is its new capability of the ice-selecting pumped counterflow virtual impactor (IS-PCVI). The IS-PCVI instrument separates ICRs from interstitial particles, including cloud droplets (*Hiranuma et al*., 2016). The IS-PCVI is a custom-built instrument that can accommodate a substantially larger counterflow as compared to commercially available PCVIs (e.g., *Boulter et al*., 2006). Such a large counterflow allows the IS-PCVI to have the critical cut-off sizes of larger than 10 μm (more than twice larger than regular PCVIs) and, therefore, inertially separate ice crystals from droplets found in mixed-phase clouds. Preserving ICRs, which are leftover INPs after evaporating water content, downstream the IS-PCVI is key to elucidate physico-chemical identities of high-*T* INPs. As described in *Hiranuma et al*. (2016), the development of the IS-PCVI was guided by computation fluid dynamics simulations, and the IS-PCVI performance was verified in the laboratory using the AIDA chamber. Verifications include its transmission efficiencies and cut-sizes up to ~30 μm, ice phase separation based on the cut-size, validation of the evaporation section as part of the IS-PCVI outlet, performance of the interstitial particle sampling and minimum artifact detection (up to 5%).

We acknowledge the AIDA technical engineering group (Rainer Buschbacher, Tomasz Chudy, Olga Dombrowski, Jens Nadolny and Georg Scheurig), hosts (Ottmar Möhler, Harald Saathoff and Nsikanabasi Umo) and the KIT-IMK-AAF science team and an IT technician (Kristina Höhler, Alexei Kiselev, Caroline Elisabeth Schaupp, Frank Schwarz, Xiaoli Shen, Isabelle Steinke, Romy Ullrich, Steffen Vogt and Robert Wagner) for maintaining and operating the cloud chamber facility as well as for data sharing. Note all names are listed in an alphabetical order.

4. Method and experimental set-up

OAF Samples: In this study, we have used four different types of OAF surface materials as surrogates for dust particles from OAFs in the Texas Panhandle. A summary of our samples is provided in **Table 1**. All samples were ground, hammer-milled and sieved for <75 μm in diameter (<10 μm for TXD02). Additionally, a heat-treated sample (i.e., 100 °C oven-dried) of each type was also examined in this study. Each sample was injected into the AIDA chamber using a rotating brush disperser (PALAS, RGB1000) followed by passing through a series of inertial cyclone impactor stages (D_{50} of ~3 μm) as demonstrated in *Hiranuma et al*. (2015) - see the **Supplement Sect. S.2** for more information regarding the OAF particle size distribution.

Table 1. Summary of four OAF samples.

ID	Description
TXD01	Raw surface material composite from several commercial and experimental feedlot surface soils
TXD02	Mechanically aerosolized <10 μm dust simulant of TXD01 generated under a concentration of <3 mg m^{-3}
TXD03	Raw surface material from a test feeding pen where cattle are fed with Tylosin (a macrolide antibiotic)
TXD05	Raw surface material from a test feeding pen where cattle are fed without antibiotics nor a direct-fed microbial (DFM) known as a probiotic

Analytical Capabilities: A total of nine techniques were used to investigate the ice-nucleating properties, in particular immersion freezing (*Vali et al*., 2015), and characterize other properties of OAF particles. As shown in **Table 2**, three techniques employed online methods that refer to measurements taken place onsite at the AIDA facility, and another set of six techniques used offline methods that denote the experiments conducted outside of the AIDA. Summary information of individual methods are provided in references given in **Table 2** and references given therein.

Table 2. Summary of instruments and techniques used in this study.

ID (Type of Characterization)	Experimental Setting	Description	Status	Reference

AIDA (IN Properties)	Online	Controlled expansion cloud-simulation chamber	Immersion freezing measurements for three different T ranges (i.e., -21.5 °C < T < -14.5 °C, -27.5 °C < T < -20.5 °C and -31.5 °C < T < -24.5 °C) all done and the analysis of IN active surface-site density, $n_s(T)$, parameterization (i.e., *Niemand et al.*, 2012) in progress	*Möhler et al.*, 2003
IS-PCVI (Chemical Properties)	Online	Inertial impactor for collecting ice residuals	Microscopy grid samples collected and measurements and analysis in progress	*Hiranuma et al.*, 2016
LAAPTOF-MS (Chemical Properties)	Online	Laser ablation aerosol particle time-of-flight mass spectrometer	Measurements all done and data analysis in progress	*Shen et al.*, 2018
INSEKT (IN Properties)	Offline	Immersion mode ice spectroemeter	Filter and <75 μm sieved-bulk samples collected and measurements and analysis in progress	*Schiebel*, 2017
DFPC (IN Properties)	Offline	Substrate-supported diffusion cell	Measurements and preliminary analysis done	*Santachiara et al.*, 2010
WT-CRAFT (IN Properties)	Offline	Cold stage-supported droplet assay	Filter samples collected and measurements and analysis in progress	*Tobo*, 2016
BET (Physical Properties)	Offline	Gas adsorption based specific surface analysis	Measurements and preliminary analysis done	*Brunauer, et al.*, 1938
Pycnometry (Physical Properties)	Offline	Gas displacement density analyzer	Measurements and analysis in progress	*Micrometrics*, 2018
Metagenomics (Biological Properties)	Offline	Total DNA extraction and sequencing of phylogenetic markers	Measurements and analysis in progress	*Jiang et al.*, 2015

5. Data description

Through 17 days of the measurement campaign, we conducted 26 AIDA expansion experiments (excluding reference expansions for the background check) and a series of supplementary experiments for LAAPTOF-MS measurements and DNA sampling for metagenomics using the aerosol particle chamber (APC). All data associated with this study (i.e., data from techniques listed in Table 2) will be archived according to the AIDA and APC experiment number. All primary data will be maintained and managed by Drs. Möhler and Ullrich at KIT. All IN data will be converted to and stored in $n_s(T)$ and/or INP concentration per volume of air at given T, $n_{INP}(T)$ (e.g., *DeMott et al.*, 2018). An example of AIDA's expansion experiment data is shown in the **Supplement Sect. S.3**. Briefly, shown in **Fig. S.2** is the time series chamber data of pressure (P), T, relative humidity (RH), aerosol concentration (C_n) and diameter of simulated-cloud particles (d). Our archived data include such a rigorous experimental data of each AIDA or APC experiment.

6. Preliminary results and conclusions

Ice Nucleation Efficiencies: Shown in **Fig. 1** is the n_s spectra of TXD01 (both pre-treated and heat-treated) in comparison to two reference spectra (i.e., U17 and O14). As seen, our OAF spectra are comparable to the previous agricultural soil dust parameterization (e.g., the n_s value of 10^{10} m^{-2} at -26 °C). More interestingly, our preliminary comparison between non-heated vs. heated samples indicates no suppression in IN ability in the T range of -21 to -28 °C. **The observed heat resistance of IN efficiency of OAF sample highlights our outcome and warrants further analyses.** Previous soil-derived INP studies (e.g., *Tobo et al.*, 2014) show a substantial effect of heat on the suppression of IN ability in this T range. Nevertheless, we will assess the influence of heat on high-T INPs by using our offline IN techniques.

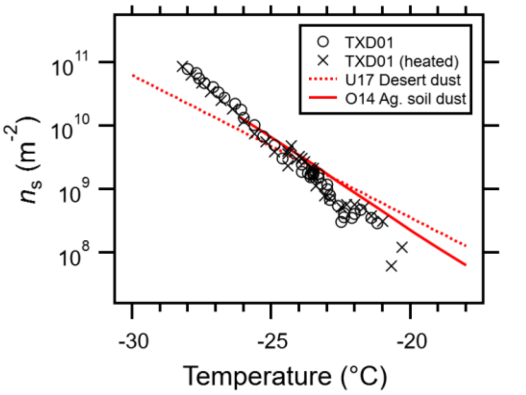

Figure 1. Ice nucleation active surface-site density, n_s, of TXD01 as a function of T. A comparison of non-heat-treated sample (circle marks; TXDUST01_07-08) to heated-sample (cross marks; TXDDUST01_03-04) is shown. Two reference lines are adapted from *Ullrich et al*. (2017; U17) and *O'Sullivan et al*. (2014; O14).

Chemical Properties: Single particle mass spectra of dry dispersed TXD01 particles in the size range between 200 and 2500 nm were measured out of APC using the LAAPTOF–MS. With the overall detection efficiency of 0.08 ± 0.05% for all the measured TXD samples (0.09% for TXD01), the LAAPTOF-MS has successfully acquired 972 spectra of TXD01. The averaged mass spectra of TXD01 are shown in **Fig. 2**. The mass spectra of the dry dispersed particles show high signals of anions at mass-to-charge ration, m/z, of 26 ($CN^-/C_2H_2^-$), 42 ($CNO^-/C_2H_2O^-$), 63 (PO_2^-) and 79 (PO_3^-). Other than that, there are typical cation markers for potassium (39 K^+) and calcium (40 Ca^+), as well as secondary organic aerosols markers (e.g., a combination of 39 $C_3H_3^+$, 41 $C_3H_5^+$; 43 $C_3H_7/C_2H_3O^+$, 44 CO_2^+ and 55 $C_4H_7/C_3H_3O^+$, and organic acids markers at 45 $COOH^-$, 59 CH_2COOH^-, 71 CCH_2COOH^-). Overall, our preliminary results of TXD01 spectra suggests the presence of four major classes, namely (1) secondary organic and phosphate rich, (2) potassium and phosphate rich, (3) potassium dominant and (4) calcium dominant class. **Interestingly, our preliminary assessment implies no notable change in chemical compositions and size distribution after heat treatment** (not shown).

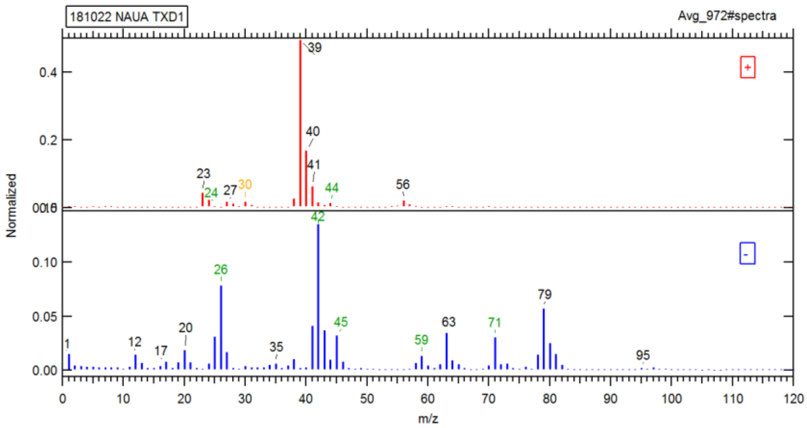

Figure 1. Average positive (top) and negative (bottom) ion mass spectra for 972 single TXD1 particles after laser ablation and ionization at 193 nm.

Physical Properties: Feedyard particles have a complex porous morphology over the volume mode of ≈10 µm in diameter (*Hiranuma et al*., 2008). **Figure 3** shows a representative scanning electron microscope (SEM) image of OAF particles. As can be seen, OAF particles surface possesses substantial amount of line structures and defects. These defects may provide a thermodynamically preferential condition to reduce the energy barrier of crystallization and thereby induce different interactions with water vapor and/or supercooled water droplets (*Page and Sear*, 2006). In addition, **Table 3** shows the preliminary results of specific surface areas and pore properties of four OAF samples, TXD01, TXD02, TXD03 and TXD05, measured by the BET-(Brunauer–Emmett–Teller) analyzer. The measured specific surface areas of OAF samples (≈2.32-3.23 $m^2\ g^{-1}$) are slightly higher compared to those of previously measured agricultural soil dust samples (0.74-2.31; *O'Sullivan et al*., 2014), but similar to that of microcline (K-feldspar; 3.2 $m^2\ g^{-1}$) and microcrystalline cellulose (3.35 $m^2\ g^{-1}$) that are known to contain the surface with substantial amount of porous structures (*Atkinson et al*., 2013; *Hiranuma et al*., 2018; *Kiselev et al*., 2017).

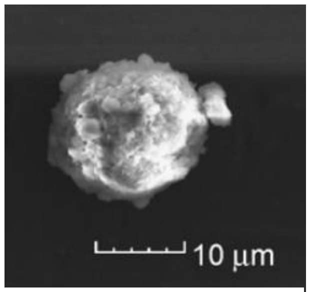

Figure 3. Representative electron microscopy image of OAF dust particles. Adapted from *Hiranuma et al*., 2008.

Table 3. Specific surface areas (SSA) of our OAF samples. All values are isotherm based using nitrogen as adsorbent. Degassing *T* was at 55 °C.

Sample	*SSA*, $m^2\ g^{-1}$
TXD01	3.233
TXD02	2.811
TXD03	2.317
TXD05	2.414

Biological Properties: Following the procedure described in *Jiang et al*. (2015), we have successfully extracted 6.74-451 ng µL^{-1} of highly pure DNA with from each sample listed in **Table 1**. Note that the extracted DNA quantity varies because DNA was extracted both from bulk dust samples as well as from dust collected using the APC-filter sample, which contains aerosolized OAF sample, before and after heating at 100 °C (a complete list of our DNA samples is available upon request). To date, our samples have been shipped for metagenomics analysis at an external, commercial lab (Eurofins Medigenomix GmbH). The metagenomics results have not been delivered yet.

7. Outlook and future studies

Future outlook and studies include:

a. Chemical properties: More detailed chemical spectral classification, size-resolved analyses and comparison to other samples are underway.

b. Physical properties: During our preliminary BET analysis, all samples were degassed at 55°C to prevent thermal degradation of biological components. We will repeat the measurements after degassing the same samples at 200 °C, to assess if the BET-SSA would increase after the volatile organics are removed. Thus, more investigation using the BET instrument is warranted. The measurement of particle density through gas-displacement pycnometry is currently in progress.

c. Biological Properties: Once the metagenomics results become available, the PIs will analyze the data for: (1) Semi-quantitative metagenomics estimation of bacterial species, number and diversity, including culturable and non-culturable bacteria, (2) Assessing species composition, such as morphology, gram stain, fluorescent pseudomonads, IN activity of isolates, (3) Identifying key bacterial and fungal/yeasts species based on selection of most prominent isolates in number, and taking into account their IN activity, and (4) Examining thermal degradation of DNA (*Karni et al*., 2013).

d. Ice Nucleation Efficiencies: Our preliminary AIDA results suggest all of our OAF samples are reasonably ice active in immersion mode freezing (not shown). More detailed and direct validation of AIDA in comparison to INSEKT, DFPC and WT-CRAFT will be conducted. Further, we will combine a parameterization of the ice-nucleating propensity of high-*T* INPs by a modern cold stage droplet freezing assay (e.g., WT-CRAFT) and ICR analyses on the residual samples collected by IS-PCVI for all of our OAF dust samples.

In succession, the data associated with this study will be made available to the modelers to incorporate the said emission flux and IN parameterization (n_s) in the atmospheric and global climate models. The PI, hosts and all participants listed will contribute to possible science journal publications.

8. References

Atkinson, J. D. et al.: Nature, 498, 355–358, 2013.
Boulter, J. E. et al.: Aerosol. Sci. Technol., 40, 969–976, 2006.
Brunauer, S. et al.: J. Am. Chem. Soc., 60, 309–319, 1938.
Bush, K. J. et al., 2014: Appl. Eng. Agric., 30, 815–824, 2014.
DeMott, P. J. et al.: Atmos. Chem. Phys., 15, 393–409, 2015.
DeMott, P. J. et al.: Atmos. Meas. Tech., 11, 6231–6257, 2018.
Després, V. R. et al.: Tellus B, 64, 15598, 2012.
Hiranuma, N. et al.: Atmos. Environ., 42, 1983–1994, 2008.
Hiranuma, N. et al.: Atmos. Chem. Phys., 11, 8809–8823, 2011.
Hiranuma, N. et al.: Atmos. Chem. Phys., 15, 2489–2518, 2015.
Hiranuma, N. et al.: Atmos. Meas. Tech., 9, 3817–3836, 2016.
Hiranuma, N. et al.: Atmos. Chem. Phys. Discuss., in review, 2018.
Hoose, C. and Möhler, O.: Atmos. Chem. Phys., 12, 9817–9854, 2012.
Jiang, W. et al.: Nat. Protoc., 10, 768–779, 2015.
Karni, M. et al.: DNA & Cell Biol., 32, 1–4, 2013.
Kiselev, A. et al.: Science, 355, 367–371, 2017.
Li, R. et al.: Sci. Rep., 7, 13826, 2017.
Micrometrics, AccuPyc™ II 1340, last access on Dec. 24, 2018, available at www.micromeritics.com/Repository/Files/AccuPyc_Final_0.pdf.
Mukhtar, S.: TCE, L-5489, 9-07, 2007.
Murray, B. J. et al.: Chem. Soc. Rev., 41, 6519–6554, 2012.
Möhler, O. et al.: Atmos. Chem. Phys., 3, 211–223, 2003.
Nagaraja, T. G.: Rumenology, Springer International Publishing, Cham, 39–61, 2016.
Niemand, M. et al.: J. Atmos. Sci., 69, 3077–3092, 2012.
O'Sullivan, D.: Atmos. Chem. Phys., 14, 1853–1867, 2014.
O'Sullivan, D.: Sci. Rep., 5, 8082, 2015.
Page, A. J. and Sear, R. P.: Physical Review Letters, 97, 065701, 2006.
Rosenfeld, D. et al.: Science, 321, 1309–1313, 2008.
Santachiara, G. et al.: Atmos. Res., 96, 266–272, 2010.
Schiebel, T.: Thesis, Karlsruhe Institute of Technology, October 20th, 2017.
Shen, X. et al.: Atmos. Meas. Tech., 11, 2325–2343, 2018.
Tobo, Y.: Atmos. Chem. Phys., 14, 8521-8531, 2014.
Tobo, Y.: Sci. Rep., 6, 32930, 2016.
USDA, 2012: last access Dec. 1, 2018, available at www.texascounties.net/statistics/cattle2012.htm.
Vali, G. et al.: Atmos. Chem. Phys., 15, 10263–10270, 2015.
Von Essen, S. G., & Auvermann, B. W.: J. Agromedicine, 10, 55–64, 2005.
Whiteside, C. et al.: AMS Conference, Austin, TX, USA, 2018, available at https://ams.confex.com/ams/98Annual/webprogram/Paper332423.html.

9. Supplement

S.1. Hypotheses to Be Tested: Our scientific hypotheses are:

1. What particulate features of OAF dust trigger IN in the high-T? This study will augment our previous field study in the Texas Panhandle (*Whiteside et al*., 2018). Biogenic aerosols in general promote nucleation of ice (*Després et al*, 2012), and they can be identified by characterizing the IN ability of heat-treated samples (at 100 °C) to non-heat-treated samples.

2. What are the INP fractions of segregated submicrometer particles (<1 μm)? Some previous size-fractionated agricultural soil dust studies suggest the presence of nano INPs that are predominantly biological fragments (<1 μm) (*O'Sullivan et al*., 2015). The predominance of submicrometer INPs suggests certain physicochemical and/or biological advantages might exist for IN in submicron particles.

3. How do ambient INP results compare to feedlot surface materials? Comparing IN ability of a surrogate of ambient OAF dust to surface soil samples aerosolized in the AIDA chamber will may shed light on long standing discussion regarding the representativeness of dried, pulverized surface materials as surrogates for ambient dust particles in immersion freezing tests.

4. How does cattle diet influence INP abundance in surface soil samples? Comparing IN ability of surface soil samples from different pens where cattle are fed with antibiotics and probiotics to those fed without antimicrobial substance may hint certain bacteria are ice-nucleation active.

5. What are contributions of OAF particle properties to its INP propensity? Because the INP number concentrations from previous field and laboratory studies increase up to 10 orders of magnitude in the heterogeneous freezing *T* range (i.e., *T* above approximately -35 °C) (e.g., DeMott et al., 2015; *Murray et al*., 2012), the OAF INP from the Texas Panhandle might exhibit a similar trend. We hypothesize that 2–3 orders of magnitude of variability at a single *T* can be explained by differences in physicochemical and/or biological properties.

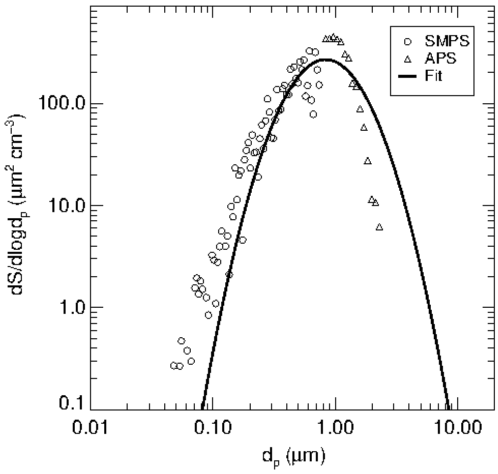

Figure S1. Surface area size distribution of TXD01 (from the experiment TXDUST01_07) measured by a combination of SMPS and APS. We assume a particle density of 1.0 g cm^{-3} and a unit dynamic shape factor.

S.2. OAF Particle Size Distribution: An example surface area size distribution of TXD01 is shown in **Fig. S1**. Prior to each expansion experiment, a combination of a scanning mobility particle sizer (SMPS, TSI, Model 3080 differential mobility analyzer and Model 3010 condensation particle counter), an aerosol particle sizer (APS, TSI, Model 3321) and a condensation particle counter (CPC, TSI, Model 3076) collectively measured the total number and size distribution of aerosols at the horizontally extended outlet of the AIDA chamber. Followed by the injection and size distribution measurement, each sample was examined for its IN ability by the expansion experiment individually.

S.3. Ice Nucleation Efficiencies: Shown in **Fig. S2** is an example of AIDA's expansion experiment profile at *Ts* between ≈-17 °C and ≈-23 °C (**Panel a**). The pressure within the chamber was reduced (Δ*P* ≈ 250 hPa), causing the *T* to drop and an adiabatic 'expansion' to occur. The AIDA chamber contained TXD01. As can be seen, measurements made by AIDA simulated immersion freezing at water saturation (**Panel b**). The time series of simulated-cloud particle concentrations (**Panels c and d**) exhibit >20 μm, indicating a formation of ice crystals.

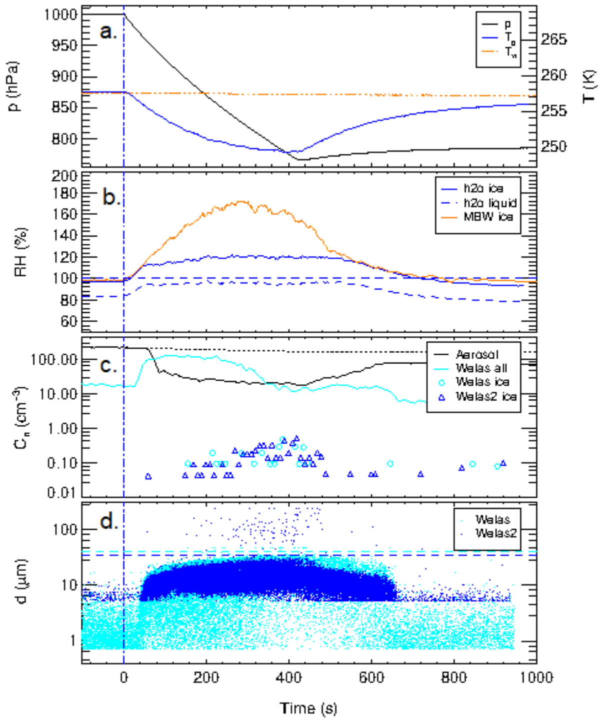

Figure S2. Experimental profile of AIDA immersion freezing experiment (TXDUST01_07).