Supplementary Information for

Feedlot is a unique and constant source of atmospheric ice-nucleating particles

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S1. Chemical Composition Analysis

- Single particle mass spectra of dry dispersed TXD particles in the size range between 200 30 and 2500 nm (vacuum aerodynamic diameter) were measured in the lab using a laser ablation aerosol particle time-of-flight mass spectrometer (LAAPTOF; AeroMegt GmbH) (Shen et al., 2018; 2019). Both untreated and heat-treated samples were examined. The powder particles were generated by powder dispersion using a rotating brush generator (PALAS GmbH, RBG1000), where small volumes of dry TXD sample were dispersed by dry synthetic air.
- 35 The averaged mass spectra of TXD01 and TXD05 are shown in **Fig. S1**. We found no significant alternations in chemical composition and size distribution after dry-heating treatment; thereby, only spectra measured for the non-treated samples are shown. Nonetheless, this negative result supports the heat tolerance of TXD particles. In general, the mass spectra of the dry dispersed particles showed high signals of organic markers at mass-to-charge ratio, m/z, of +44
- 40 (COO/C₂H₆N⁺), -26 (CN/C₂H₂⁻), -42 (CNO/C₂H₂O⁻), -45 (COOH⁻), -59 (CH₂COOH⁻), -71 (CCH₂COOH⁻), +30 (NO/CH₃NH/CH₂O⁺), +58 (C₂H₅-NH-CH₂⁺), and +59 ((CH₃)₃N⁺). These are typical markers for organic acids and amine-containing particles. For example, peaks at m/z of +44 can be attributed to COO/CH2NO⁺ derived from organic compounds/nitrogen containing organic compounds (Schneider et al., 2011). It should be noted that m/z 44 can also be contributed by
- SiO⁺, which is silicon marker (Silva and Prather, 2000). Further, -45 (COOH⁻), -59 (CH₂COOH⁻), and -71 (CCH₂COOH⁻) are the markers for carboxylic acids. Peak at m/z of +30 can be attributed to NO⁺ arising from nitrate, ammonium (Murphy et al., 2006; Shen et al., 2018), and CH₃NH⁺ from amines (Silva and Prather, 2000; Schmidt et al., 2017). The other amine markers at +58 (C₂H₅NHCH₂⁺) and +59 ((CH₃)₃N⁺) were identified by previous studies (e.g., Angelino et al., 2001; Pratt et al., 2009; Schmidt et al., 2017).
- 50 Pratt et al., 2009; Schmidt et al., 2017). For the inorganic markers, the characteristic ions were found on the peaks at m/z +23 (Na⁺), +24 (Mg⁺), +27 (Al⁺), +28 (Si⁺), +39 (K⁺), +40 (Ca⁺), +44 (SiO⁺), +56 (CaO/Fe⁺), +64/66 (Zn⁺), -97 (HSO₄⁻), +30 (NO⁺), -63 (PO₂⁻), -79 (PO₃⁻), and -95 (PO₄⁻). Calcium and sodium are used as additives in the diet fed to the cattle, and they also exist in the unpaved road dust (National Research Council, 2000; Ocsay et al., 2006). Manure is a source of ammonium and phosphate.

Minor fractions of other salts and mineral dust constituents, found in this work, were also identified in the field samples (Hiranuma et al., 2011 and references therein). As mentioned above, +30 NO⁺ can arise from ammonium (Murphy et al., 2006; Shen et al., 2018). In addition, -63 (PO₂), -79 (PO₃), and -95 (PO₄) are phosphate markers (Schmidt et al., 2017; Zawadowicz et al., 2017). However, our inorganic quantification is inconclusive, and the result may deviate from other

quantitative composition analyses.

Comparing TXD01 to TXD05, we found that TXD01 had more intensive phosphate (-63, -79) and potassium (+39) compared to TXD05 (**Fig. S1**). In particular, phosphate intensity was a few times higher than TDX05. On the other hand, TXD05 had a higher signals of sodium- and nitrogen-containing compounds as well as stronger amine markers, i.e., m/z +30 (NO/CH₃NH⁺) and +58 (C₂H₅-NH-CH₂⁺), than TXD01.

A more detailed analysis of the individual mass spectra revealed several distinct particle types. Using a combination of the fuzzy c-means clustering (Shen et al., 2019) and the marker peak search method based on the above-mentioned and other characteristic ions, we found several

- 15 distinct composition classes, such as "Potassium rich," "Potassium and phosphate rich," "Potassium, sodium, and ammonium rich," "Amine rich," and "Mineral and Metal rich." We note that the "rich" used here only indicates intensive characteristic peaks in the mass spectra rather than a large mass fraction. Figure S2 shows the fuzzy classification results. As can be seen, there was no notable size dependent composition for any sample types. No obvious change in chemical
- 20 compositions and size distribution was found after dry-heating treatment. A slight decrease in organic-potassium mixtures was found for dry-heated particle samples in comparison to non-heated ones, but the difference was insignificant. A significant amount of carboxylic acid groups (i.e., m/z -45 and -71) was found in each particle. These prevalent organic markers suggest that, regardless of the classification, TXD are organic-predominant in nature. This organic predominance
- 25 as well as substantial inclusion of salts (e.g., potassium) are consistent with our previous study of TXD particles composition (Hiranuma et al., 2011). We also note that our LAAPTOF aerosol particle chemical composition analysis was not intended to find IN active composition. Ice-nucleating particles generally represent a small subset of aerosol particles (roughly one per million, even at low *T*s). Thus, examining aerosol particle chemical composition cannot be directly linked to the role
- 30 of chemistry in IN. In other words, aerosol particle composition does not necessarily represent icenucleating particle composition. A complementary elemental composition of ice crystal residuals is discussed in the main manuscript. But, aerosol particle composition data are important for understanding the general chemical compositions of our samples.



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Figure S1. Laboratory reference mass spectra of dry dispersed TXD01 and TXD05 particles with LAAPTOF. The left panels show the stacked averaged spectra of cations (top) and anions (bottom) found in TXD01 and TXD05. The right panels represent the absolute signal difference. These mass spectra represent a compilation of > 450 of the particles for each type (TXD01: 972 and TXD05: 472). Note that each ion peak intensity is normalized to the sum of ion signal in each spectrum before further compilation.



Figure S2. Particle population fraction and size distribution based on clustered types, for TXD01 (a), TXD01H (b), TXD05 (c), and TXD05H (d). Note that the class named "others" (in grey color) is the small fraction of particles with unknown patterns. This class differs across TXD particle samples.

S2. Comparison of two immersion freezing techniques. As shown in Fig. S3, the West Texas Cryogenic Refrigerator Applied to Freezing Test system (WT-CRAFT) system measured the immersion mode freezing efficiency of a bulk test open-air feedlot material (TXD01). This complementary analysis was meaningful to indirectly validate WT-CRAFT against the Ice Nucleation SpEctrometer of the Karlsruhe Institute of Technology (INSEKT) measurements. The

- 5 Nucleation SpEctrometer of the Karlsruhe Institute of Technology (INSEKT) measurements. The comparison to INSEKT showed that the ice nucleation (IN) active surface site density values, $n_s(T)$, over -8°C > Temperature (T) > -21°C agreed within an order of magnitude. Further, both methods successfully captured a local maximum in freezing spectra shown in **Fig. S3** at around -10°C, suggesting that they are comparable immersion freezing detection techniques. The uncertainty of INSEKT with respect to Tis + 0.5°C (Schiebel, 2017)
- 10 INSEKT with respect to T is ± 0.5 °C (Schiebel, 2017).



Figure S3. Immersion freezing spectra of the bulk TXD01b sample measured by KIT-INSEKT and WT-CRAFT. This figure shows the BET surface-scaled $n_s(T)$ spectra (Hiranuma et al., 2015). For the ice-nucleating particle measurement uncertainty, 95% binomial sampling confidence intervals (CI95%), are computed using the procedure described in Schiebel (2017, Eqn. 3.21). The vertical error bars represent CI95%.

S3. Taxonomic diversity of two Texas dust samples. We examined the diversity of *Archaea*, *Bacteria*, and *Fungi* in TXD01 and TXD05. Bulk samples, dry-heated bulks (100°C for 12 hours), and aerosolized particles collected on nuclepore filters were analyzed. Useful data for *Bacteria*

- 20 were generated from the amplification and sequencing of the V3-V5 region of the 16S rDNA phylogenetic marker down to the genus and species level. The predominant phyla of *Archaea* consisted of methanogens, colonizers of the bovine rumen, as expected (Fouts et al., 2012) (**Table S1a**). The bacterial fraction of the microbiome was dominated by *Actinobacteria* (the most abundant phylum, common soil inhabitants), *Proteobacteria, Firmicutes* (diverse bacterial phyla with species
- 25 living in the soil as well as in the bovine rumen), and Bacteroidetes (common members of the bovine rumen microflora) (Fouts et al., 2012; Chaucheyras-Durand and Ossa, 2014). No known IN active bacterial species were identified in either sample (Després et al., 2012), although the genus *Pseudomonas* (containing IN active species) was detected in low numbers (**Table S1b**). The predominant fungal taxa in our samples belong to *Pezizomycetes* (*Ascomycota*), common soil
- 30 inhabitants. In this taxon, the coprophilic genus Ascobolus was detected in high numbers, as expected (Sarrocco, 2016). The genera Fusarium (Ascomycota-Hypocreales) and Mortierella (Mucuromycota-Mortierellales) were also detected in low numbers. These genera contain species with IN activity; however, the phylogenetic analysis did not detect any known IN active species of these genera (Table S1c). Nonetheless, it is very interesting that we did not identify any known IN
- 35 active biological species in our samples.

Table S1. Abundance of major orders of *Archaea* in dust samples TXD01 and TXD05 (**a**). Numbers indicate percentage of the OTUs for each phylum in the total archaeal microbiome. The analysis of the aerosolized TXD01 sample and of dry heat-treated bulk samples did not generate any useful data. Abundance of major bacterial and eukaryotic (incl. fungal) orders in dust samples TXD01 and TXD05 are shown in (**b**) and (**c**). Numbers indicate percentage of the OTUs for each order in the total bacterial and eukaryotic microbiome.

a. Archaea Taxonomy	Bulk TXD01	Bulk TXD01 (Dry- heated)	Aerosolized TXD01	Bulk TXD05	Bulk TXD05 (Dry- heated)	Aerosolized TXD05
Unclassified	4.10%	-	-	0.00%	-	0.00%
Euryarchaeota; Methanobacteria; Methanobacteriales	58.40%	-	-	77.70%	-	93.80%
Euryarchaeota; Methanomicrobia; Methanomicrobiales	18.30%	-	-	1.50%	-	0.10%
Euryarchaeota; Methanomicrobia; Methanosarcinales	0.60%	-	-	5.00%	-	0.00%
Euryarchaeota; Thermoplasmata; Methanomassiliicoccales	12.30%	-	-	0.00%	-	0.00%
Thaumarchaeota; Nitrososphaeria; Nitrososphaerales	6.30%	-	-	15.80%	-	6.10%
b. Bacteria Taxonomy	Bulk TXD01	Bulk TXD01 (Dry- heated)	Aerosolized TXD01	Bulk TXD05	Bulk TXD05 (Dry- heated)	Aerosolized TXD05
Unclassified	3.80%	4.00%	4.00%	8.10%	8.60%	2.60%
Actinobacteria: Acidimicrobiales	1.50%	1.50%	0.80%	1.40%	1.20%	0.20%
Actinobacteria; unclassified	0.50%	1.20%	3.00%	4.70%	6.60%	2.10%
Actinobacteria; Actinomycetales	0.30%	0.30%	0.20%	0.10%	0.00%	0.00%
Actinobacteria; Bifidobacteriales	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Actinobacteria; Corynebacteriales	11.10%	11.40%	16.40%	14.00%	12.90%	13.70%
Actinobacteria; Frankiales	0.50%	0.30%	0.20%	0.00%	0.00%	0.00%
Actinobacteria; Geodermatophilales	0.40%	0.60%	0.30%	0.00%	0.00%	0.00%
Actinobacteria; Glycomycetales	0.00%	0.00%	0.20%	0.00%	0.30%	0.40%
Actinobacteria; Jiangellales	0.10%	0.10%	0.00%	0.10%	0.20%	0.00%
Actinobacteria; Kineosporiales	0.00%	0.10%	0.00%	0.00%	0.00%	0.00%
Actinobacteria; Micrococcales	20.10%	20.10%	12.30%	13.00%	12.20%	2.10%
Actinobacteria; Micromonosporales	0.00%	0.00%	0.10%	0.00%	0.10%	0.00%
Actinobacteria; Propionibacteriales	5.60%	5.90%	5.00%	3.90%	2.70%	0.20%
Actinobacteria; Pseudonocardiales	0.00%	0.00%	7.70%	6.70%	14.30%	39.20%
Actinobacteria; Streptomycetales	0.20%	0.60%	11.30%	5.60%	11.90%	28.60%
Actinobacteria; Streptosporangiales	0.00%	0.00%	2.30%	1.70%	4.70%	6.50%
Actinobacteria; Coriobacteriales	0.00%	0.00%	0.00%	0.10%	0.00%	0.00%
Actinobacteria; Solirubrobacterales	0.00%	0.00%	0.20%	0.40%	0.50%	0.00%
Bacteroidetes; unclassified	0.10%	0.10%	0.10%	0.80%	0.50%	0.00%
Bacteroidetes; Chitinophagales	0.70%	0.60%	0.30%	0.00%	0.00%	0.00%
Bacteroidetes; Cytophagales	1.60%	2.50%	0.70%	0.10%	0.20%	0.00%
Bacteroidetes; Flavobacteriales	8.60%	8.60%	4.20%	0.10%	0.00%	0.00%
Bacteroidetes; Saprospirales	0.20%	0.10%	0.10%	0.00%	0.00%	0.00%
Bacteroidetes; Sphingobacteriales	1.50%	1.30%	1.10%	1.30%	0.90%	0.00%
Chloroflexi; Sphaerobacterales	4.40%	4.90%	4.00%	9.30%	4.00%	1.10%
Cyanobacteria; Chroococcales	0.30%	0.10%	0.00%	0.00%	0.00%	0.00%
Fibrobacteres; Fibrobacterales	0.20%	0.00%	0.00%	0.00%	0.00%	0.00%
Firmicutes; unclassified	0.10%	0.10%	0.10%	0.00%	0.00%	0.00%
Firmicutes; Bacilli; unclassified	0.00%	0.00%	0.10%	0.10%	0.10%	0.00%
Firmicutes; Bacillales	11.90%	12.40%	6.10%	5.30%	3.90%	2.40%
Firmicutes; Lactobacillales	1.50%	1.20%	0.60%	1.50%	0.60%	0.00%

Firmicutes; Clostridiales	3.20%	3.40%	5.90%	6.90%	1.80%	0.30%
Firmicutes; Erysipelotrichales	0.40%	0.40%	1.00%	1.50%	0.40%	0.10%
Firmicutes; Acidaminococcales	0.00%	0.00%	0.00%	0.10%	0.00%	0.00%
Firmicutes; Tissierellia; unclassified	0.00%	0.00%	0.00%	0.10%	0.00%	0.00%
Firmicutes; Tissierellales	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Gemmatimonadetes;	0.80%	0.90%	0.40%	0.00%	0.00%	0.00%
Gemmatimonadales	0.8078	0.90%	0.40%	0.0078	0.0078	0.00%
Gemmatimonadetes; Longimicrobiales	0.80%	0.50%	0.00%	0.10%	0.00%	0.00%
Nitrospinae; Nitrospinales	0.10%	0.00%	0.00%	0.00%	0.00%	0.00%
Planctomycetes; Candidatus Brocadiales	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Proteobacteria; unclassified	0.40%	0.30%	0.10%	0.50%	0.20%	0.00%
Proteobacteria; Alphaproteobacteria; unclassified	0.30%	0.30%	0.30%	0.00%	0.10%	0.00%
Proteobacteria; Alphaproteobacteria; Caulobacterales	1.10%	1.20%	0.50%	0.00%	0.00%	0.00%
Proteobacteria; Alphaproteobacteria; Rhizobiales	4.40%	2.90%	2.90%	1.00%	1.40%	0.00%
Proteobacteria; Alphaproteobacteria; Rhodobacterales	1.30%	1.60%	0.50%	0.50%	0.30%	0.00%
Proteobacteria; Alphaproteobacteria; Rhodospirillales	0.00%	0.10%	0.00%	0.00%	0.00%	0.00%
Proteobacteria; Alphaproteobacteria; Sphingomonadales	2.60%	3.30%	1.60%	0.10%	0.20%	0.00%
Proteobacteria; Betaproteobacteria; Burkholderiales	0.80%	0.70%	1.30%	2.00%	3.60%	0.00%
Proteobacteria; Deltaproteobacteria; Desulfuromonadales	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Proteobacteria; Deltaproteobacteria; Myxococcales	0.00%	0.00%	0.00%	0.00%	0.10%	0.00%
Proteobacteria; Gammaproteobacteria: unclassified	0.10%	0.00%	0.00%	0.30%	0.00%	0.00%
Proteobacteria; Gammaproteobacteria; Aeromonadales	0.00%	0.00%	0.00%	0.10%	0.00%	0.00%
Proteobacteria; Gammaproteobacteria; Cardiobacteriales	0.10%	0.00%	0.00%	0.00%	0.00%	0.00%
Proteobacteria; Gammaproteobacteria; Cellvibrionales	3.40%	2.40%	0.40%	0.00%	0.20%	0.00%
Proteobacteria; Gammaproteobacteria; Chromatiales	0.10%	0.00%	0.00%	0.00%	0.00%	0.00%
Proteobacteria; Gammaproteobacteria; Enterobacterales	0.00%	0.00%	1.60%	6.10%	4.00%	0.50%
Proteobacteria; Gammaproteobacteria; Nevskiales	0.20%	0.10%	0.00%	0.00%	0.00%	0.00%
Proteobacteria; Gammaproteobacteria; Oceanospirillales	0.00%	0.00%	0.00%	0.10%	0.10%	0.00%
Proteobacteria; Gammaproteobacteria; Pseudomonadales	0.70%	0.30%	0.60%	1.80%	1.00%	0.00%
Proteobacteria; Gammaproteobacteria; Xanthomonadales	2.80%	2.20%	1.00%	0.30%	0.20%	0.00%
Proteobacteria; Bdellovibrionales	1.20%	1.40%	0.50%	0.10%	0.00%	0.00%
Rhodothermaeota; Rhodothermales	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Spirochaetes; Spirochaetales	0.00%	0.00%	0.00%	0.10%	0.00%	0.00%

		Bulk TXD01			Bulk TXD05	
c. Eukaryotic Taxonomy	Bulk TXD01	(Dry- heated)	Aerosolized TXD01	Bulk TXD05	(Dry- heated)	Aerosolized TXD05
Unclassified	18.10%	19.20%	0.30%	0.50%	0.00%	1.60%
Trichiida	0.70%	1.30%	0.00%	0.00%	0.30%	0.00%
Oligohymenophorea; Philasterida	5.10%	0.90%	0.00%	0.00%	0.00%	0.00%
Oligohymenophorea; Sessilida	0.30%	5.40%	0.00%	0.00%	0.00%	0.00%
Phyllopharyngea; Chlamydodontida	0.10%	0.40%	0.00%	0.00%	0.00%	0.00%
Spirotrichea; Sporadotrichida	0.90%	0.00%	0.00%	0.00%	0.00%	0.00%
Ascomycota; unclassified	0.60%	0.80%	1.10%	1.40%	0.00%	0.90%
Ascomycota; Capnodiales	0.70%	0.50%	0.00%	0.00%	1.10%	0.00%
Ascomycota; Pleosporales	2.10%	0.70%	0.00%	0.00%	0.00%	0.00%
Ascomycota; Eurotiales	0.10%	1.40%	1.30%	2.40%	0.00%	2.80%
Ascomycota; Onygenales	0.00%	0.00%	1.70%	1.40%	1.20%	5.20%
Ascomycota; Pertusariales	0.30%	0.00%	0.00%	0.00%	1.80%	0.00%
Ascomycota; Leotiomycetes; unclassified	6.10%	0.40%	0.10%	0.00%	0.00%	0.00%
Ascomycota; Rhytismatales	0.00%	6.90%	0.00%	0.00%	0.00%	0.00%
Ascomycota; Thelebolales	1.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Ascomycota; Pezizales	46.90%	1.50%	68.00%	64.60%	0.00%	20.40%
Ascomycota; Saccharomycetales	0.50%	45.90%	0.10%	0.20%	65.90%	0.10%
Ascomycota; Glomerellales	0.20%	0.10%	0.00%	0.00%	0.10%	0.00%
Ascomycota; Hypocreales	0.30%	0.20%	16.90%	16.90%	0.00%	59.50%
Ascomycota; Melanosporales	0.00%	0.20%	0.10%	0.00%	18.80%	0.10%
Ascomycota; Microascales	2.40%	0.00%	0.60%	2.10%	0.10%	3.10%
Ascomycota; Sordariales	9.20%	2.30%	5.30%	3.70%	0.80%	2.80%
Basidiomycota; unclassified	0.00%	6.60%	0.00%	0.00%	5.80%	0.00%
Basidiomycota; Sporidiobolales	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Basidiomycota; Tremellomycetes; unclassified	0.20%	0.00%	0.00%	0.00%	0.00%	0.00%
Basidiomycota; Trichosporonales	0.00%	0.20%	4.40%	6.40%	0.00%	3.30%
Basidiomycota; Wallemiales	0.00%	0.00%	0.00%	0.00%	4.10%	0.00%
Chytridiomycota; Rhizophlyctidales	1.00%	0.00%	0.00%	0.00%	0.00%	0.00%
Chytridiomycota; Spizellomycetales	1.60%	1.40%	0.00%	0.00%	0.00%	0.00%
Chytridiomycota; Neocallimastigales	1.00%	1.90%	0.00%	0.00%	0.00%	0.00%
Mucoromycota; Mortierellales	0.50%	1.30%	0.00%	0.00%	0.00%	0.00%
Mucoromycota; Mucorales	0.10%	0.50%	0.10%	0.40%	0.00%	0.20%

SI References

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- S. Angelino, D. T. Suess, K. A. Prather, Formation of aerosol particles from reactions of secondary and tertiary alkylamines: Characterization by aerosol time-of-flight mass spectrometry. *Environ. Sci. Technol.* **35**, 3130–3138 (2001).
- F. Chaucheyras-Durand, F. Ossa, REVIEW: The rumen microbiome: Composition, abundance, diversity, and new investigative tools. *The Professional Animal Scientist.* **30**, 1–12 (2014).
- V. R. Després et al., Primary biological aerosols in the atmosphere: A review of observations and relevance. *Tellus B.* **64**, 15598 (2012).
 - D. E. Fouts et al., Next generation sequencing to define prokaryotic and fungal diversity in the bovine rumen. *PLoS One.* **7**, e48289 (2012).
- N. Hiranuma, S. D. Brooks, J. Gramann, B. W. Auvermann, High concentrations of coarse particles emitted from a cattle feeding operation. *Atmos. Chem. Phys.* **11**, 8809–8823 (2011).

- N. Hiranuma et al., A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques. *Atmos. Chem. Phys.* **15**, 2489–2518 (2015).
- D. M. Murphy et al., Single-particle mass spectrometry of tropospheric aerosol particles. J. *Geophys. Res.-Atmos.* **111**, D23S32 (2006).
- National Research Council. "Minerals" in *Nutrient Requirements of Beef Cattle: Seventh Revised Edition: Update 2000* (The National Academies Press, 2000), pp. 54–69.
- R. Ocsay, I. Salma, W. Wang, W. Maenhaut, Characterization and diurnal variation of sizeresolved inorganic water soluble ions at a rural background site. *J. Environ. Monitor.* **8**, 300–306 (2006).
- K. A. Pratt, L. E. Hatch, K. A. Prather, Seasonal volatility dependence of ambient particle phase amines. *Environ. Sci. Technol.* **43**, 5276–5281 (2009).
- S. Sarrocco, Dung-inhabiting fungi: a potential reservoir of novel secondary metabolites for the control of plant pathogens. *Pest. Manag. Sci.* **72**, 643–652 (2016).
- T. Schiebel, Ice nucleation activity of soil dust aerosols. Thesis, Karlsruhe Institute of Technology, https://doi.org/10.5445/IR/1000076327 (20 October 2017).
 - S. Schmidt et al., Online single particle analysis of ice particle residuals from mountain-top mixed-phase clouds using laboratory derived particle type assignment. *Atmos. Chem. Phys.* **17**, 575–594 (2017).
- J. Schneider et al., Mass- spectrometric identification of primary biological particle markers and application to pristine submicron aerosol measurements in Amazonia. *Atmos. Chem. Phys.* **11**, 11415–11429 (2011).
 - X. Shen et al., Laser ablation aerosol particle time-of-flight mass spectrometer (LAAPTOF): Performance, reference spectra and classification of atmospheric samples. *Atmos. Meas. Tech.* **11**, 2325–2343 (2018).
 - X. Shen et al., Understanding of atmospheric aerosol particles with improved particle identification and quantification by single particle mass spectrometry. *Atmos. Meas. Tech.* **12**, 2219–2240 (2019).
 - P. J. Silva, R. A. Carlin, K. A. Prather, Single particle analysis of suspended soil dust from Southern California. *Atmos. Environ.* **34**, 1811–1820 (2000).
 - M. A. Zawadowicz, K. D. Froyd, D. Murphy, D. J. Cziczo, Improved identification of primary biological aerosol particles using single particle mass spectrometry. *Atmos. Chem. Phys.* 17, 7193–7212 (2017).

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