# **1. Supplementary Methods**

## 2 1.1 DNA-Containing Cell Enumeration

3	Cells in the precipitation were preserved by adding sodium borate-buffered
4	formalin (pH=8.2, stored at room temperature) to a final concentration of 5% v/v.
5	Triplicate samples from each precipitation event were processed by filtering 10 mL of
6	sample onto $0.22\mu m$ , 25mm black polycarbonate filters (Millipore) and staining with a
7	final concentration of 25X SYBR-Gold (Invitrogen) for 15 min. in the dark. Cell density
8	estimates were obtained using an epifluorescence microscope (Olympus bx51) and data
9	from 60 fields of view (1 field of view=34636 $\mu$ m <sup>2</sup> ).
10	
11	1.2 DNA Extraction, Sequencing, and Analysis
12	The thawed 47mm Supor PES membrane filters were transferred to a laminar
13	flow hood and cut into small pieces using sterile scissors. The filter pieces were
14	transferred to a bead beating tube from the FastDNA <sup><math>TM</math></sup> SPIN Kit for Soil (MP
15	Biomedicals, Santa Ana, CA). The DNA was extracted according to the manufacturer's
16	protocol, with the following modifications: in step 4, a mini bead beater was used to
17	homogenize extracts for 70 seconds; step 5 was performed for 8 minutes; a 15 mL tube
18	was used for step 7; and 100 $\mu$ L of DNase/pyrogen-free water was used to elute DNA in
19	step 16. The extracts obtained were further purified using steps 14-22 of the
20	manufacturer's protocol for the MoBio Power Soil Kit (MoBio Laboratories, Carlsbad,
21	CA). In these steps, 160 $\mu L$ of solution C4 was added to the DNA extraction. The
22	manufacturer's protocol was followed for all other steps with the exception of adding 25

23	$\mu$ L of solution C6 and incubating at room temperature prior to centrifugation. The DNA
24	was stored at $-20$ °C prior to polymerase chain reaction (PCR) amplification.
25	The V4 region of the bacterial 16S rRNA gene was PCR amplified in triplicate
26	from each DNA extract for sequencing on the Illumina MiSeq (Illumina, Inc., San Diego,
27	CA, USA). The PCR was carried out using the barcoded primers and methods of
28	Caporaso et al. (1). The $25\mu L$ reaction contained the following components: 5 Prime
29	Master Mix (1X), 0.5 $\mu M$ 515F, 0.5 $\mu M$ 806R and nuclease free water (13 $\mu L)$ and 2.0
30	$\mu$ L of DNA. Thermal-cycling was carried out in an Eppendorf PRO S Master Cycler
31	(Eppendorf North America, Hauppauge, NY, USA) under the following conditions:
32	initial denaturation at 94 °C for 3 min. followed by 32 cycles of denaturation at 94 °C for
33	1 min., annealing at 50 °C for 1 min. extension at 72 °C for 1 min. 45 s, and followed by
34	a final extension at 72 $^{\circ}$ C for 10 min. The amplicons obtained were evaluated by
35	electrophoretic separation on a 1% agarose gel buffered with Tris-acetate-EDTA and
36	stained with ethidium bromide. PCR products from the triplicate reactions were pooled,
37	purified, and concentrated using the Qiagen MinElute PCR Cleanup kit (Qiagen,
38	Valencia, California, USA) with the optional 35% guanidine-HCl wash step to ensure
39	removal of large primer-dimers. Amplicons were stored at $-20$ °C until sequencing at
40	Idaho State University's Molecular Research Core Facilty (Pocatello, ID, USA) on the
41	Illumina MiSeq Platform (Illumina Inc., San Diego, CA) using the V2 500 bp kit.
42	Sequence data were analyzed using the mothur software package (2). Contigs
43	were assembled and parsed on the basis of unique barcodes attached to the 806R primer
44	(1). Sequences that did not contain exact matches to the primer and barcodes utilized in
45	the PCR amplification were discarded. Sequences were filtered for quality with a 50-base

47	ambiguous bases, homopolymers (> 7 bases), or having lengths > 259 bases were
48	eliminated from the dataset.
49	
50	1.3 Inorganic Chemical Analyses
51	Conductivity and pH were measured using a multi-parameter PCSTest probe
52	(Oakton Instruments, Vernon Hills, IL). Samples of deionized water were routinely
53	analyzed and served as procedural blanks. A Dionex ICS-3000 ion chromatography
54	system was used to determine the concentration of major ions in the precipitation
55	samples. The system was equipped for anion separation with a 4×250 mm RFIC <sup>TM</sup>
56	IonPac® AS18 column (Dionex Corporation, CA, USA), using a water:potassium
57	hydroxide eluent, and a 4×250 mm RFIC <sup>™</sup> IonPac <sup>®</sup> CS16 column (Dionex Corporation,
58	CA, USA) with a water: methanesulfonic acid eluent. Guard columns (Dionex
59	Corporation, CA, USA) preceded each column (4×50 mm RFIC <sup>TM</sup> IonPac <sup>®</sup> AG18 guard
60	column was used for the anion channel and a 4×50 mm RFIC <sup>™</sup> IonPac <sup>®</sup> CG16 guard
61	column for the cation channel). Each sample was analyzed in triplicate. With this method,
62	2 carboxylic acids (formate and oxalate) and 13 inorganic ions (F <sup>-</sup> , Cl <sup>-</sup> , NO <sup>2-</sup> , Br <sup>-</sup> , NO <sup>3-</sup> ,
63	$SO_4^{2-}$ , $PO_4^{3-}$ , $Li^+$ , $Na^+$ , $NH_4^+$ , $K^+$ , $Mg^{2+}$ and $Ca^{2+}$ ) could be quantified. The limit of
64	detection, calculated as three times the standard deviation of the field blanks, was
65	between 0.1 to 0.8 $\mu$ M for all ions reported.
66	
67	1.4 Organic Carbon Concentration, Fluorescent Characterization, and PARAFAC

sliding window and a minimum average quality score of 25, and those containing

68 Modeling

69 DOC concentrations were obtained from a GE Sievers 900 Total Organic Carbon 70 Analyzer. An average DOC measurement was calculated from three measurements of 71 organic carbon concentrations for each precipitation filtrate sample (sample size ~25 72 mL). Blank samples of Milli-Q Water were measured between each sample to monitor 73 successive sample-to-sample contamination throughout instrument use. Acidification 74 was not necessary prior to experimentation due to an internal acidification step within the 75 instrument.

76 A Horiba Jobin Yvon Fluoromax-4 Spectrofluorometer generated the Excitation 77 Emission Matrices (EEMs) of the fluorescent dissolved organic matter (DOM) in the 78 precipitation samples. This instrument is equipped with a Xenon lamp light source and a 79 1 cm path length quartz cuvette was used for all measurements. Excitation (Ex) 80 wavelengths were scanned from 240-450 nm in 10 nm intervals and emission (Em) was 81 recorded between 300-560 nm in 2 nm increments. Data integration time was 0.25 s and 82 data acquisition was carried out in signal/reference mode using a 5 nm bandpass on both 83 Ex and Em monochromators, normalizing the fluorescence Em signal with the Ex light 84 intensity. Absorbance spectra (190-1100nm) was incorporated into the spectral correction 85 calculations of primary and secondary inner filter effects for post-processing the 86 fluorescence data to generate EEMs (3, 4). Spectra were blank corrected against purified 87 water from a Milli-Q system each day. A Parallel factor analysis (PARAFAC) model 88 was generated in MATLAB by drEEM and the N-way toolbox scripts (5) to determine 89 individual DOM fluorescing components in the EEMs.

90

91 1.5 Meteorological Data Collection and Analysis

92	On-site meteorological data was collected continuously with a weather station
93	(Vantage Pro, Davis Instruments), and supplemented with data obtained locally through
94	the Louisiana Agriclimatic Information System (automated weather station at Ben Hur
95	Agricultural Fields, ~5 km SE of the main sampling location).
96	Storm classification was based on cloud top data retrieved from The National
97	Weather Service (NWS) archive of Geostationary Operational Environmental Satellite-
98	East (GOES-East) infrared and visible satellite imagery and Next Generation Radar
99	(NEXRAD) Level III radar reflectivity provided by the National Climatic Data Center's
100	(NCDC) website (https://www.ncdc.noaa.gov/nexradinv/). Troposphere temperature
101	profiles were retrieved from The NWS radiosonde data archive of stations located in
102	Lake Charles, Louisiana (LCH, station number 72240) and Slidell Muni, Louisiana (LIX
103	station number 72233).

104 Classification of convective and nimbostratus precipitation based on radar 105 reflectivity and satellite imagery was carried out according to previous methods (6, 7). 106 Tropospheric stability indices from NWS soundings (8, 9) were used to confirm the 107 presence or lack of Convective Available Potential Energy (CAPE), which indicates the 108 presence of convection. Convection occurs when the surface of the earth is heated 109 unevenly, leading to the warming of air directly above the heated surface. This warmer air 110 is more buoyant than the surrounding air and begins to rise. Once this "parcel" of warm air 111 rises to the "convective condensation level" (CCL), water vapor will begin to condense 112 and form water droplets, and subsequently, a cloud develops. If precipitation came from a 113 cloud which was formed in the presence of convection the Convective Condensation Level 114 (CCL) was used to estimate the height of the cloud base (Supplementary Figure S1 a).

115 Convergence (Supplementary Figure S1 b) occurs when a low pressure system is present. 116 Air in high pressure regions moves towards lower pressure regions, leading to the 117 convergence of air masses, forcing the air to move up in the atmosphere. Warm front 118 (Supplementary Figure S1 c) lifting occurs when a warm front advances and the less dense, 119 warm air within that warm front is displaced upward over cooler, denser air ahead of it. 120 Cold front (Supplementary Figure S1 d) lifting occurs when a cold front advances and 121 displaces the warmer air ahead of it upward. Orographic lifting was not observed in this 122 study and is not depicted. If precipitation came from a cloud which was formed in the 123 absence of convection and by one of the prior three methods listed, the Lifted Condensation 124 Level (LCL) was used to estimate the height of the cloud base (Supplementary Figure S1 125 b-d).

126 As an example of how trajectories were analyzed, panels f and e in Supplementary 127 Figure S1 show the altitudes and trajectories used for a particular rain event that occurred 128 on August 25, 2014. The cloud base for this event was at approximately 1160 mAGL, and 129 the cloud top was at approximately 16800 mAGL. Given that the cloud system developed 130 through convection (Supplementary Figure S1 a), the six altitudes chosen (depicted in 131 panels a-d as the gray dotted lines) for HYSPLIT backward trajectory analysis were *below* 132 the cloud at the time of precipitation in Baton Rouge. The six backward trajectories were 133 then examined for previous interactions with the MBL or surface using the tdump csv files 134 generated by HYSPLIT, in which trajectory height, MBL height, and surface height are 135 listed at hourly intervals for each trajectory. If at any point along the trajectory history 136 (histories of which ranged from 120-168 hours) the air masses descended into the MBL or 137 interacted with the ground, the geographic coordinates these interactions were recorded for that event and plotted in R to determine the corresponding ecoregion (Supplementary Figure S1 f). Panel f shows the geographic coordinates of the trajectories for this precipitation event where interactions with the surface of MBL occurred. These coordinates were then mapped to the ecoregions outlined in Figure 1. Trajectory and ecoregion interactions are listed in Supplementary Dataset S1.

143 Cloud top heights were estimated using the Equilibrium Levels (EL) and
144 Maximum Parcel Levels (MPL), in addition to NCDC's Level III echo top data, which

estimates cloud height based on recorded pressure and temperature levels. Herein,

stratiform precipitation is defined specifically as precipitation that was collected from

147 stratus and nimbostratus-like cloud systems independent of trailing stratiform regions

148 from convective storm formations (10).

149 Once cloud formation type was determined, six unique altitudes were chosen to

150 be analyzed for 120-168 h backward trajectory analysis based on the CCL or LCL of the

151 precipitation event (Supplementary Figure S1a-d). Trajectories were analyzed for

152 previous interactions with the surface and/or mixed boundary layer (MBL)

153 (Supplementary Figure S1e). This was accomplished by downloading the "tdump.csv"

154 files produced by HYSPLIT, which detailed the recorded height above ground level of

the trajectory being analyzed, in addition to the height of the MBL.

156

157 *1.6 Statistical Analyses* 

158 The statistical procedures (Exploratory Factor Analysis, Multiple Imputation,

159 Analysis of Variance, Multivariate Analysis of Variance, Mann-Whitney U-Test,

160 Welch's Tests, Kruskall-Wallis Test, Pearson's and Spearman's Correlations, and

161 Tukey's Honest Significant Difference post-hoc analysis) were performed using SAS

162 software, Version 9.4 of the SAS System for Windows. Graphs and plots were produced

using R Software Version 3.2.1 (The R Core Team 2015).

164 Prior to hypothesis testing with analysis of variance (ANOVA) and multivariate 165 analysis of variance (MANOVA), the raw data were screened for univariate and 166 multivariate normality using The Shapiro-Wilk Test and through visual inspection of Q-167 Q plots. The assumption of homoscedasticity was verified using Levene's test, univariate 168 outliers were examined based on z-score distributions, multivariate outliers identified 169 using Mahalanobis distance, and linearity/collinearity evaluated through visual inspection 170 of bivariate scatter plots. Log and arcsine transformations were used on distributions that 171 violated assumptions of normality. For distributions not corrected by data 172 transformations, hypothesis tests that assume non-normality or heteroscedasticity were 173 used (i.e., Mann-Whitney U-test, Kruskall-Wallis one-way ANOVA, and Welch's Test).

174 For extreme outliers (values more than three times the interquartile range), raw data

175 values were adjusted according to the method of Tabachnick and Fidell (11).

176 Multiple imputation was used to provide missing INP concentration data prior to

177 Exploratory Factor Analysis (EFA) and hypothesis testing. The differential INP

178 concentrations were grouped and summed based on the results of the EFA. For example,

the INP concentrations used to represent Bio<sub>-5 to -10</sub> is the summed differential

180 concentrations of INPs active between -5 and  $-10^{\circ}$ C.

181 For hypothesis testing, the dependent variables were always the summed

182 differential INP concentrations for each INP category determined by EFA, which were

183 continuous variables. When the independent variables were continuous (Cell abundance,

- 184 pH, conductivity, major ion concentrations, DOC concentrations, PARAFAC
- 185 Components C1-C3 intensities, OTU sequence reads), Pearson's R and Spearman's Rank
- 186 correlational analyses were used. Note that for correlations between INP concentrations
- and bacterial taxa, the number of OTU sequence reads was used for the analysis. When
- 188 the independent variables were categorical (ecoregion classification, cloud type, season,
- and precipitation type), ANOVA and MANOVA were used. For post-hoc analysis of
- 190 ANOVA and MANOVA, Tukey's Honest Significant Difference (HSD) analysis was
- 191 used. Tukey's HSD test is a post-hoc analysis that compares the means of each group to
- 192 find significant differences between groups (11).
- 193

# 194References195

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227 228		
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## 232 Supplementary Table S1. Results of Multiple Imputation for missing INP data. Missing data

column lists the variables which did not contain an observation. All missing data followed a

- 234 monotone missing data pattern.

Missing data	Sample	Missing Data Pattern	Number of	Relative	$\mathbf{Pr} >  \mathbf{t} ^{\mathcal{C}}$
	Size	<b>Frequency</b> <sup><i>a</i></sup>	Imputations	<b>Efficiency</b> <sup>b</sup>	
			Performed	-	
total INPs					
≤-13°C	4	6.56%	5	0.98	< 0.0001
≤-14°C	8	13.11%	5	0.92	< 0.0001
-15°C	3	4.92%	5	0.92	< 0.0001
<u>biological</u>					
INPs					
≤-13°C	6	9.84%	5	0.95	< 0.0001
≤-14°C	6	9.84%	5	0.93	< 0.0001
-15°C	4	6.56%	5	0.89	< 0.0001
bacterial INPs					
≤-13°C	3	13.46%	5	0.98	< 0.0001
≤-14°C	7	13.46%	5	0.92	< 0.0001
-15°C	7	5.77%	5	0.95	< 0.0001

<sup>a</sup>The percent of observations containing the specified missing data

<sup>b</sup>Measure of how well the imputation calculations converged, as described in Li et al., JAMA 314:1966–1967, 2015.

 $^{c}$ P-value for t-test of H<sub>0</sub>: mean=0

- 263 Supplementary Table S2. MANOVA results of INP concentrations, interactions of air
- 264 masses, and ecoregions. PM, Pacific Maritime; NAM, North Atlantic Maritime; SAM,
- 265 South Atlantic Maritime; NFM, Northwest Forested Mountains; DSAH, Desert and
- 266 Semi-Arid Highlands; HNL, High Northern Latitudes; GP, Great Plains; EWW, Eastern
- 267 Woodlands and Wetlands; EA, East Asia.

INP class	Ecoregions of	significance <sup>a</sup>	ANOVA test results			
	<u>Highest INP</u> concentrations <sup>a</sup>	<u>Lowest INP</u> concentrations <sup>a</sup>	Prob > F			
total <sub>-5 to -11</sub>	EA, HNL	NAM, SAM	P < 0.0001			
total <sub>-11 to -14</sub>	EA	NAM, SAM, EWW, DSAH	P < 0.001			
bio <sub>-5 to -10</sub>	EA, HNL	NAM, SAM	P < 0.0001			
bio-13 to -14	EA	NAM, SAM, EWW, DSAH	P < 0.0001			
bio_11 to -12	NFM	NAM	P < 0.05			
bac-5 to -10	EA, HNL	SAM, EWW, DSAH	P < 0.001			
<sup>a</sup> Based on significant differences in INP concentrations between ecoregions determined via						
Tukey-Kramer HSD Connecting Letters Report (MANOVA post-hoc analysis) at a confidence level of alpha=0.05						

#### Supplementary Table S3. Characteristics of fluorescent dissolved organic matter

#### PARAFAC components in precipitation from air masses interacting with distinct

ecoregions. Ecoregions are based on Level 1 Ecoregions defined by the EPA and CEC. The

PARAFAC component means are shown as Raman Units. Numbers following ecoregion name

correspond to numbers listed in the ecoregion column of Supplementary Dataset S1.

298 299	Ecoregion	Avg. Fluorescence Intensity Maximum
300		C1: 0.0035
301	Pacific Maritime (1)	C2: 0.0072
302		C3: 0.0017
202		
303	North Atlantic Maritime	N/A
304	(3)	
305		
306		C1: 0.0036
307	South Atlantic Maritime	C2: 0.0062
308	(4)	C3: 0.0040
309		
310	Northwest Forested	C1: 0.0041
311	Mountains (5)	C2: 0.0065
312		C3: 0.0035
313		C1: 0.0043
21/	Desert and Semi-Aric	C2: 0.0066
21F	Highlands (0)	C3: 0.0037
315 216	High Northern Latitudes	C1: 0.0071
310	(7)	C2: 0.0083
317	(1)	C3: 0.0032
318		C1: 0.0035
319	Great Plains (8)	C2: 0.0053
320		C1: 0.0013
321	Eastern Woodlands and	$C_{1}^{2} = 0.0041$
322	Wetlands (9)	C3: 0.0002
323		C1: 0.0071
324	East Asia (11)	C2: 0.0083
325		C3: 0.0032

- **Supplementary Table S4.** Results of multivariate analysis of variance (MANOVA) for
- all INP (total, biological, and bacterial) concentrations as a function of season, cloud
- 337 type, and precipitation type.

Meteorological Parameter	MANOVA Test Results
Season	F(15, 122) = 3.12 <b>p</b> = <b>0.0003</b>
Cloud Type	F(5, 46) = 2.47 <b>p=0.0462</b>
Precipitation Type	F(5, 46) = 2.45 <b>p=0.0472</b>
Bolded <i>p</i> -values indicate statistically significate whole Model, F Test, Prob>F	ant differences between air masses tested at alpha=0.05 lev

### 354 Supplementary Table S5. Correlations between ice nucleating particle (INP) factors and

**local meteorological conditions.** Pearson correlation coefficients (*r*) calculated between INP

356 factors and locally recorded meteorological data. Relative humidity (RH%). Significance levels

of Pearson correlation coefficients: \*p < .05, \*\*p < .01, \*\*\*p < .001. Cloud top temperature,

358 surface temperature, surface wind speed N=61; Relative humidity, rain amount N=60.

INP Factor	Cloud top temperature (°C)	Surface Temperature (°C)	RH %	Rain Amount (mm h <sup>-</sup> )	Surface wind speed (mph)
<u>Total</u>					
total <sub>-5 to -11</sub>	.07	53***	24	.06	.45***
total <sub>-11 to -14</sub>	.01	.30*	08	18	17
<b>Biological</b>					
bio <sub>-5 to</sub> -10	.06	53***	24	.05	.45***
bio-13 to -14	05	01	03	07	12
bio-11 to -12	.18	-0.29*	005	14	.04
Bacterial					
bac-5 to -10	01	-0.51***	30*	12	.32*

385 Supplementary Table S6. Significant Spearman's rank correlation coefficients (for which rho

386  $\rho \ge 0.40$ ; and significance p<0.05) between ice nucleating particle (INP) factors and taxon

abundance. Significance levels of Spearman's rank correlation coefficients: \*p < .05, \*\*p < .01,

- 388 \*\*\*p < .001. Only taxa with relative abundance >0.1% for total number of sequence reads across
- all precipitation events were analyzed. Total number of sequence reads across all precipitation
- 390 events are listed in last column.

	total-5 to -11	total-11 to -14	<b>bio</b> -5 to -10	<b>bio</b> -11 to -12	<b>bio</b> -13 to -14	bac-5 to -10	No. Sequence Reads
Taxon (Order; Family; Genus)	Spearma n's rho $(\rho)$	Spearman's rho $(\rho)$	Spearman's rho ( $\rho$ )	Spearma n's rho $(\rho)$	Spearm an's rho $(\rho)$	Spearman's rho $(\rho)$	
Acidobacteria							
Acidobacteriales; Acidobacteriaceae; Candidatus Chloracidobacterium			0.41*				5674
Bacteroldetes							
Bacteroidales; Rikenellaceae; N/A	0.62***	0.43*	0.59***		0.55***	0.54**	81
Sphingobacteriales; N/A; N/A	0.51**		0.53**				67269
Cytophagales; Cyclobacteriaceae; N/A		0.40*					514
Cytophagales; Cyclobacteriaceae; Algoriphagus				0.45*			69
Cytophagales; Cytophagaceae; N/A	0.51**		0.55***			0.43*	28658
Cytophagales; Hymenobacteraceae; Hymenobacter	0.56***	0.42*	0.59***			0.48**	19161
Cytophagales; Cytophagaceae; Spirosoma			0.41*				4486
Chitinophagales; Chitinophagaceae; Segetibacter	0.55***	0.45*	0.53**		0.44*	0.55**	294
Sphingobacteriales; env.OPS_17; N/A	0.52**	0.45*	0.44*		0.52**	0.46*	672
Cytophagales; Cytophagaceae; Flexibacter	0.42*	0.53**	0.44*	0.48**			277
Sphingobacteriales; Sphingobacteriaceae; N/A	0.41*		0.40*				20330

	total-5 to	total-11 to	<b>bio</b> -5 to -10	bio-11 to	<b>bio</b> -13 to	bac-5 to -10	No.
	-11	-14		-12	-14		Sequence Reads
Candidate Division					0.42*		81
Chlorobi							
Chlorobiales; N/A; N/A	0.42*						144
Cyanobacteria			0.40*				707954
Firmicutes							
Bacillales; Planococcaceae:		0.53**					888
Planococcus							
Bacillales; Staphylococcaceae;		0.57***			0.41*		271
Macrococcus							
Clostridiales; Lachnospiraceae; Blautia	0.43*	0.56***	0.43*				75
Clostridiales; Lachnospiraceae; Roseburia		0.41*					53
Erysipelotrichales; Erysipelotrichaceae; Turicibacter		0.47**					679
Lactobacillales; Carnobacteriaceae; N/A	0.47**	0.49**	0.46*			0.40*	642
Lactobacillales; Carnobacteriaceae; Carnobacterium	0.41*	0.48**	0.43*				157
Lactobacillales; Carnobacteriaceae; Desemzia			0.40*				91
Lactobacillales; Lactobacillaceae: N/A		0.42*					16067
Lactobacillales; Leuconostocaceae; Leuconostoc					0.47**		457
Planctomycetes							
Planctomycetales; Planctomycetaceae; Planctomyces		0.47**					105
Proteobacteria							
Campylobacterales; Campylobacteraceae; Arcobacter		0.45*		0.43*			300
Bacteriovoracales; Bacteriovoraceae; N/A		0.45*					770
Bacteriovoracales; Bacteriovoraceae; Peredibacter		0.47**					730

	total-5 to	total-11 to	<b>bio</b> -5 to -10	<b>bio</b> -11 to	<b>bio</b> -13 to	bac-5 to -10	No.
	-11	-14		-12	-14		Sequence Reads
Rhizobiales;				0.45*			523
Methylocystaceae; Methylosinus							
Sphingomonadales;					0.41*		1190
<i>Erythrobactereaceae</i> ; N/A							
Oceanospirillales;		0.44*			0.42*		66
Oceanospirillaceae;							
N/A		0.41*					2012
Pseudomonadales;		0.41*					2013
Moraxellaceae; Perlucidibaca							
Rurkholderiales:						0.42*	108
Comamonadaceae:						0.42	100
Polaromonas							
Chromatiales;		0.50*					912
Chromatiaceae;							
Rheinheimera							
Rhodocyclales;	0.47**				0.52**	0.48*	8105
<i>Rhodocyclaceae;</i> N/A				0.40.5			1001
Rhodospirillales; wr0007; N/A				0.40*			1094
Xanthomonadales;	0.47**	0.44*	0.49**				14094
Xanthomonadaceae;							
N/A Spirachaotos							
		0.41*			0.42*		02
Spirocnaetales; N/A;		0.41*			0.43*		93
Verrucomicrobia							
Chthaniahastanalast					0.42*		510
Chinoniobacterales; Chthoniobacteraceae:					0.42*		510
Chinomobacteraceae, Chthoniobacter							
Unclassified							
Unclassified: OTU20						0.40*	425
Unclassified; OTU32						0.42*	175
Unclassified; OTU43					0.43*		1020
Unclassified: OTU51				0.43*			1051
Unclassified; OTU73	0.52**	0.45*	0.44*		0.52**	0.46*	351
Unclassified; OTU74				0.42*			672
Unclassified; OTU88				0.44*			254
Unclassified; OTU108					0.46*		120
Unclassified: OTU13	0.47*		0.52**				232
Unclassified: OTU18					0.43*		146
Unclassified: OTU22	0.44*		0.51**			 	110
Unclassified, UTU22	0.44		0.51				110

## 395 Supplementary Table S7. Taxa with significantly different abundances based on cloud type

and season. Table lists the test performed (Mann-Whitney U-Test (MT), Welch's T-Test (WT), Kruskall-Wallis ANOVA (KA), and Welch's ANOVA (WA)) and corresponding *p*-value. Only significant *p*-values (p<0.05) are shown. The taxa analyzed are listed in Supplementary Table S6.

399	· · · ·	2	11 7
400	Taxon (Order; Family;		G
401	Genus)	Cloud Type	Season
402	Bacteroidetes		
402	Cytophagales;	WT 0.0291	WA 0.0112
403	Cytophagaceae; N/A		
404	Cytophagales;	WT 0.0469	
405	Cytophagaceae; Flexibacter		
406	Cytophagales;	WT 0.0231	WA 0.0108
407	Hymenobacteraceae;		
408	Hymenobacter		
409	Bacteroidales;	WT 0.0384	
410	Rikenellaceae; N/A		
411	Sphingobacteriales;	MT 0.0393	WA 0.0209
412	Sphingobacteriaceae; N/A		
413	Sphingobacteriales; N/A;	MT 0.0056	WA 0.0029
414	N/A		
415	Firmicutes		
416	Clostridiales;	MT 0.0005	
417	Lachnospiraceae; Blautia		
418	Lactobacillales;	WT 0.0233	WA 0.0400
A10	Carnobacteriaceae; N/A		
419	Lactobacillales;	WT 0.0003	
420	Carnobacteriaceae;		
421	Carnobacterium		
422	Planctomycetes	1	
423	Planctomycetales;		WA 0.0353
424	Planctomycetaceae;		
425	Planctomyces		
426	Proteobacteria		
427	Campylobacterales;	MT 0.0031	
428	Campylobacteraceae;		
429	Arcobacter		
430	Rhodocyclales;		WA 0.0111
431	<i>Rhodocyclaceae;</i> N/A		
432	Verrumicrobia		W4.0.0226
433	Chthoniobacterales;	MI 0.0118	WA 0.0326
434	Chthoniobacteraceae;		
131	Chinoniobacter		
136	Unclassified	MT 0 0099	KA 0 0007
427	Unclassified; OTU20	MT 0.0088	KA 0.0007
420 420	Unclassified; OTU32	MT 0.0452	WA 0.0299
430	Unclassified; OTU/4	WT 0.0344	
439	Unclassified; OTU15	WT 0.0278	WA 0.0049
440	Unclassified; UTU18	W I U.U391 MT 0.0187	
441	Unclassified; UTU22	M1 0.018/	

## 442 Supplementary Table S8. Spearman correlations between ice nucleating particle (INP)

443 factors. Correlations calculated for differential concentrations of INPs between factor groupings.

444 Significance levels of Pearson correlation coefficients (top number in each cell) and Spearman's

445 rho (bottom number in each cell): \*p < .05, \*\*p < .01, \*\*\*p < .001.

	total-5 to -11	total-11 to -14	<b>bio</b> -5 to -10	<b>bio</b> -13 to -14	<b>bio</b> -11 to -12	bac-5 to -10
total <sub>-5 to</sub> -11						
<b>total</b> -11 to -14	.08 .03					
<b>bio</b> –5 to –10	.96*** .94***	.20 .16				
<b>bio</b> -13 to -14	01 07	.47** .62***	.00 .01			
<b>bio</b> -11 to -12	.04 02	.53** .55***	.00 05	.00 .06		
bac-5 to -10	.83*** .83***	.26 .32*	.84*** .83***	.06 .07	.10 .15	





472 Supplementary Figure S1. Cloud formation mechanisms and HYSPLIT trajectory analysis. 473 The mechanism of lifting for each precipitation event is important in determining the altitudes for 474 backward trajectory analysis. There are five general mechanisms of large-scale air movement that 475 lead to cloud formation, which are dealt with as described in the supplemental methods. (a) 476 Convection (b) Convergence (c) Warm front lifting (d) Cold front lifting. Orographic lifting was 477 not observed in this study and is not depicted. Panels e and f show an example of how trajectories 478 were analyzed, and are described in detail in the supplemental methods. Trajectory and ecoregion 479 interactions are listed in Supplementary Dataset S1. 480



483
484 Supplementary Figure S2. PARAFAC Components fluorescence intensity profiles based on The
485 North American Ecoregion classifications used in this study. Average Fluorescence Intensity was

486 calculated based on ecoregion and is plotted on the y-axis in Raman Units (R.U.). PARAFAC

487 Components C1-C3 are plotted as categories on the x-axis.



Supplementary Figure S3. Significant differences in DNA operational taxonomic unit (OTU) abundances as a function of cloud type and season. The mean number of sequence reads for each OTU is plotted, with bars indicating the standard error of the mean. Each taxon is represented by a single unique OTU. Top: OTUs that correlated with ice nucleating particle (INP) concentrations and had significantly different abundances in precipitation from stratiform (N=10) and convective (N=35) cloud formations. Bottom: OTUs that correlated with INP concentrations and had significantly different abundances based on season (Autumn, N=12; Spring, N=6; Summer, N=14; Winter, N=13).