1. Supplementary Methods

1.1 DNA-Containing Cell Enumeration

68 *Modeling*

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

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

1.5 Meteorological Data Collection and Analysis

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

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

 As an example of how trajectories were analyzed, panels f and e in Supplementary Figure S1 show the altitudes and trajectories used for a particular rain event that occurred on August 25, 2014. The cloud base for this event was at approximately 1160 mAGL, and the cloud top was at approximately 16800 mAGL. Given that the cloud system developed through convection (Supplementary Figure S1 a), the six altitudes chosen (depicted in panels a-d as the gray dotted lines) for HYSPLIT backward trajectory analysis were *below* 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 generated by HYSPLIT, in which trajectory height, MBL height, and surface height are listed at hourly intervals for each trajectory. If at any point along the trajectory history (histories of which ranged from 120-168 hours) the air masses descended into the MBL or 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.

 Cloud top heights were estimated using the Equilibrium Levels (EL) and 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,

146 stratiform precipitation is defined specifically as precipitation that was collected from

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

148 from convective storm formations (10).

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

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

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

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

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

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.

1.6 Statistical Analyses

The statistical procedures (Exploratory Factor Analysis, Multiple Imputation,

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

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

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

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).

 Prior to hypothesis testing with analysis of variance (ANOVA) and multivariate analysis of variance (MANOVA), the raw data were screened for univariate and multivariate normality using The Shapiro-Wilk Test and through visual inspection of Q- Q plots. The assumption of homoscedasticity was verified using Levene's test, univariate outliers were examined based on z-score distributions, multivariate outliers identified using Mahalanobis distance, and linearity/collinearity evaluated through visual inspection of bivariate scatter plots. Log and arcsine transformations were used on distributions that violated assumptions of normality. For distributions not corrected by data transformations, hypothesis tests that assume non-normality or heteroscedasticity were used (i.e., Mann-Whitney U-test, Kruskall-Wallis one-way ANOVA, and Welch's Test). For extreme outliers (values more than three times the interquartile range), raw data values were adjusted according to the method of Tabachnick and Fidell (11). Multiple imputation was used to provide missing INP concentration data prior to Exploratory Factor Analysis (EFA) and hypothesis testing. The differential INP concentrations were grouped and summed based on the results of the EFA. For example, 179 the INP concentrations used to represent Bio- $5\text{ to }-10$ is the summed differential

180 concentrations of INPs active between -5 and -10° C.

For hypothesis testing, the dependent variables were always the summed

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

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

- pH, conductivity, major ion concentrations, DOC concentrations, PARAFAC
- Components C1-C3 intensities, OTU sequence reads), Pearson's R and Spearman's Rank
- 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
- the independent variables were categorical (ecoregion classification, cloud type, season,
- and precipitation type), ANOVA and MANOVA were used. For post-hoc analysis of
- ANOVA and MANOVA, Tukey's Honest Significant Difference (HSD) analysis was
- used. Tukey's HSD test is a post-hoc analysis that compares the means of each group to
- find significant differences between groups (11).
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References

232 **Supplementary Table S1. Results of Multiple Imputation for missing INP data.** Missing data

233 column lists the variables which did not contain an observation. All missing data followed a 234 monotone missing data pattern.

- monotone missing data pattern.
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^{*a*}The percent of observations containing the specified missing data

*b*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 H₀: mean=0

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- **Supplementary Table S2.** MANOVA results of INP concentrations, interactions of air
- masses, and ecoregions. PM, Pacific Maritime; NAM, North Atlantic Maritime; SAM,
- South Atlantic Maritime; NFM, Northwest Forested Mountains; DSAH, Desert and
- Semi-Arid Highlands; HNL, High Northern Latitudes; GP, Great Plains; EWW, Eastern
- Woodlands and Wetlands; EA, East Asia.
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292 **Supplementary Table S3. Characteristics of fluorescent dissolved organic matter**

293 **PARAFAC components in precipitation from air masses interacting with distinct**

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

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

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

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

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

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

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

357 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.

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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

387 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
- 389 all precipitation events were analyzed. Total number of sequence reads across all precipitation
- 390 events are listed in last column.

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

396 **and season.** Table lists the test performed (Mann-Whitney U-Test (MT), Welch's T-Test (WT), 397 Kruskall-Wallis ANOVA (KA), and Welch's ANOVA (WA)) and corresponding *p*-value. Only 397 Kruskall-Wallis ANOVA (KA), and Welch's ANOVA (WA)) and corresponding *p*-value. Only
398 significant *p*-values (*p*<0.05) are shown. The taxa analyzed are listed in Supplementary Table S6 significant *p*-values (p <0.05) are shown. The taxa analyzed are listed in Supplementary Table S6.

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): $*p < .05, **p < .01, ***p < .001$.

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PARAFAC Components

 Supplementary Figure S2. PARAFAC Components fluorescence intensity profiles based on The North American Ecoregion classifications used in this study. Average Fluorescence Intensity was calculated based on ecoregion and is plotted on the y-axis in Raman Units (R.U.). PARAFAC Components C1-C3 are plotted as categories on the x-axis.

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 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) 501 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).

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