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Supporting Information for

Deciphering the significant role of biological ice nucleators in precipitation at the organic molecular level

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Contents of this file

Introduction

This supporting information includes figures and tables.

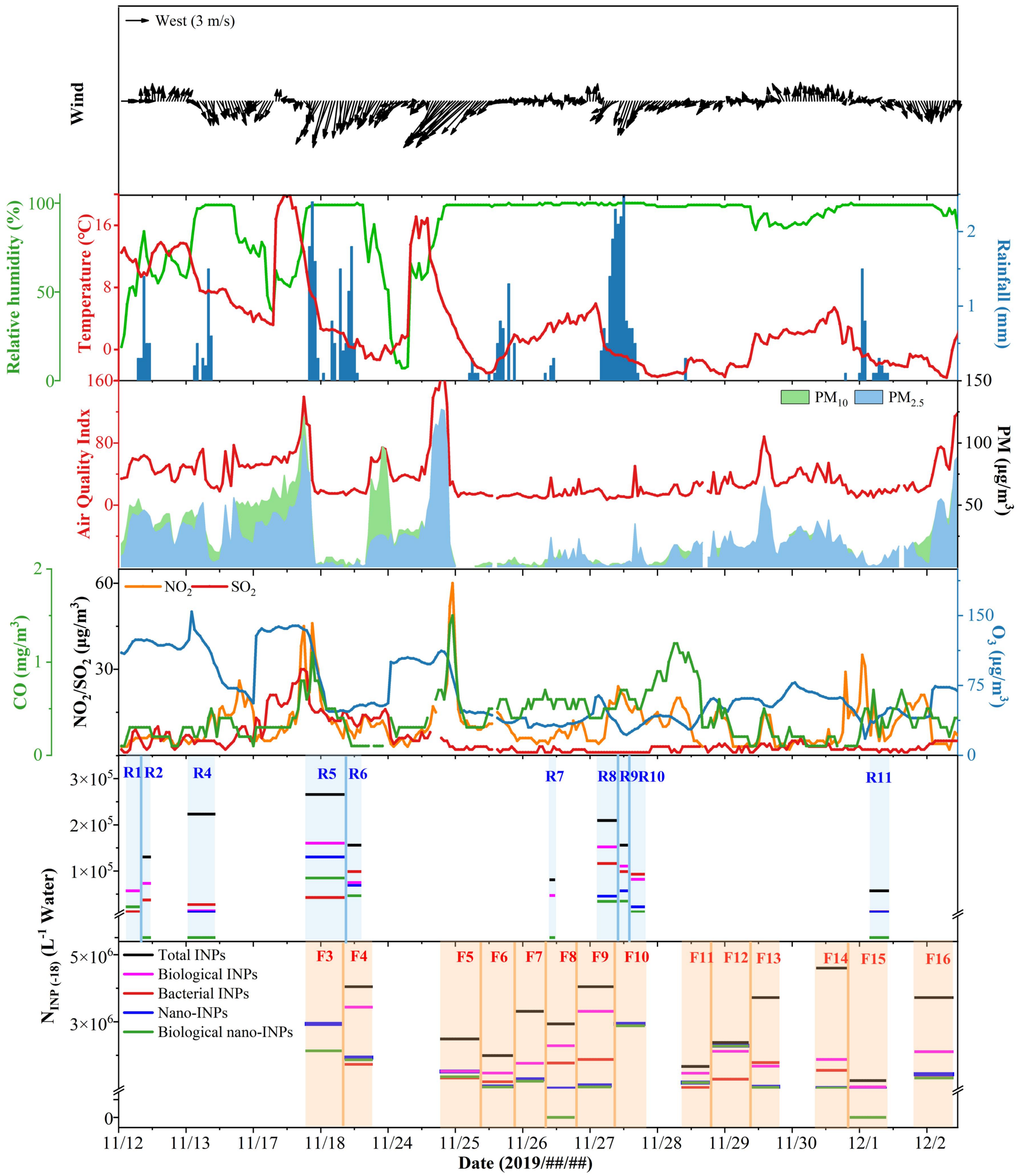

Figure S1. Data of environmental parameters (wind speed, wind direction, air temperature, relative humidity, rainfall, particulate matter (PM), NO_2, SO_2, O_3 and CO) during the sampling period and the concentrations of different types of INPs. For the sample IDs, F and R denote fog water and rainwater samples, respectively. F1, F2, and R3 were omitted from analyses due to small precipitation amounts.

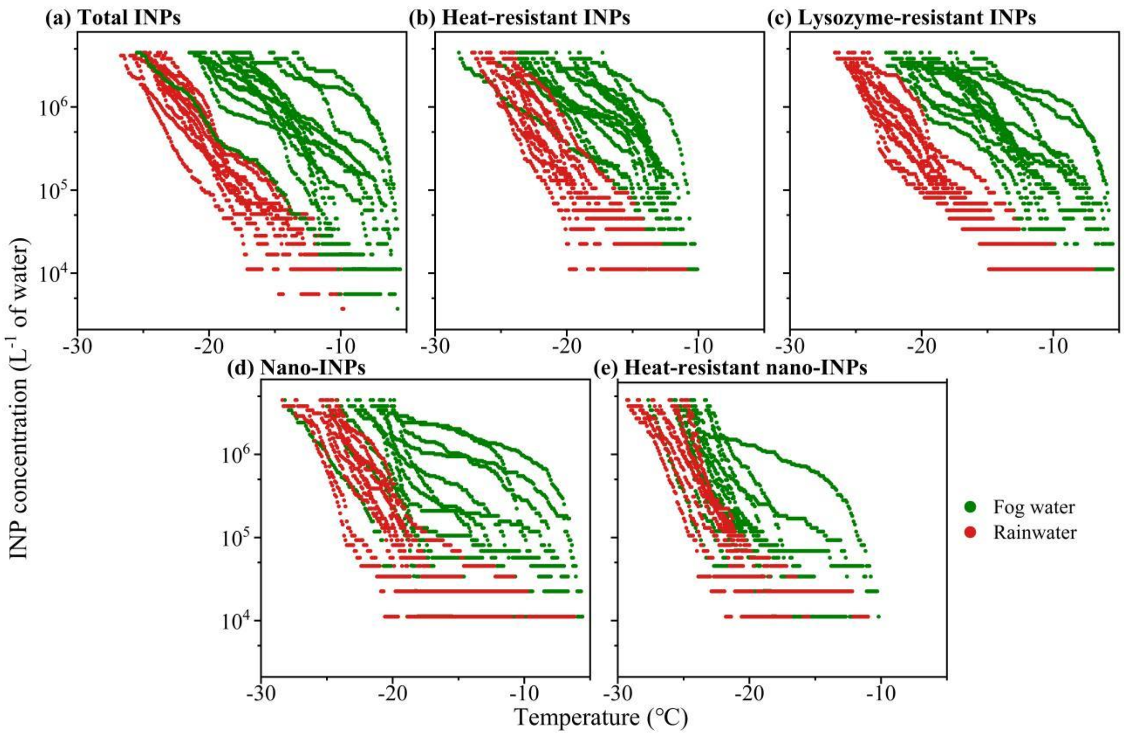

Figure S2. Comparisons of the total ice nucleating particles (INPs) spectra with heat-resistant INP spectra, lysozyme-resistant INPs spectra, nano-INPs spectra, and heat-resistant nano-INPs spectra obtained from rainwater samples (red dots) and fog water samples (green dots).

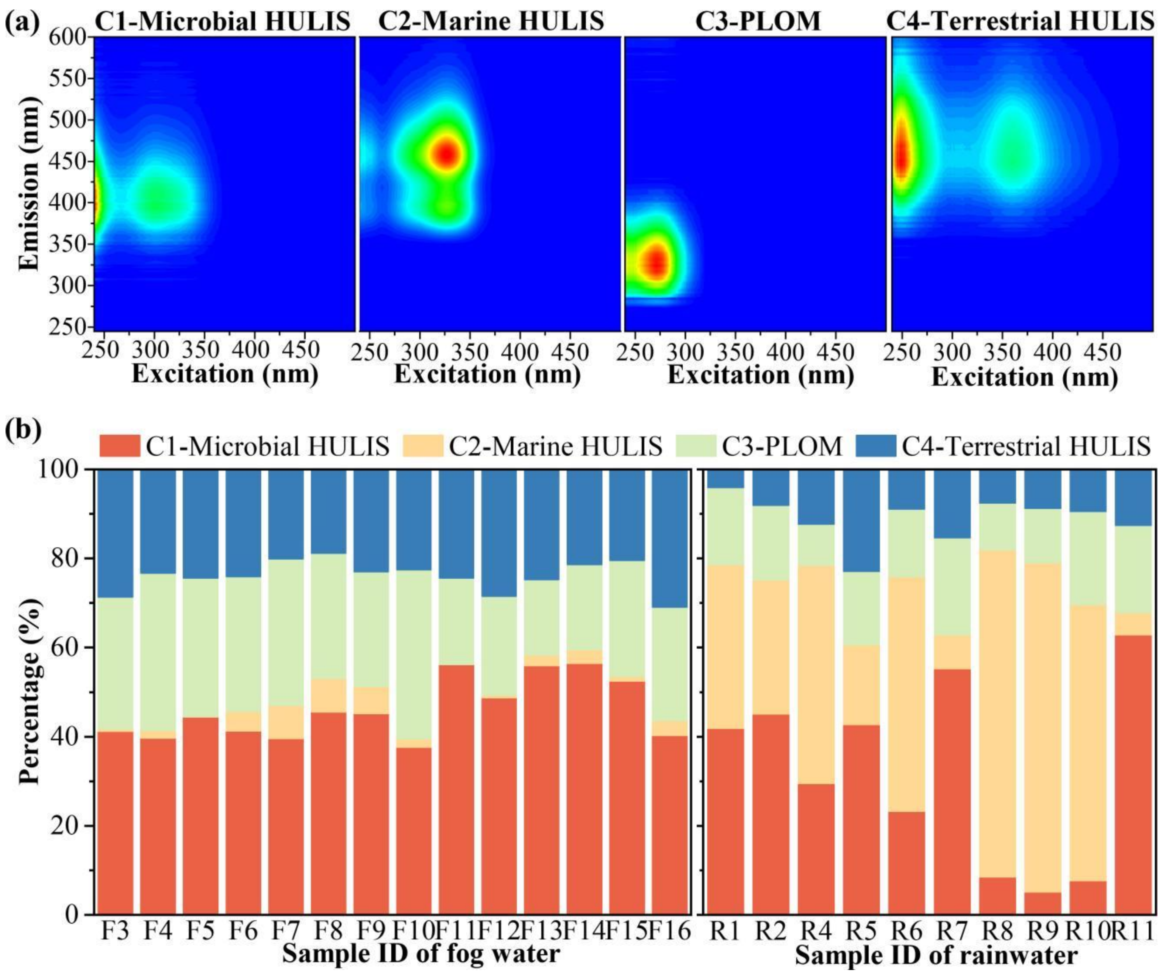

Figure S3. Fluorescence signatures of four components (C1-C4) and their percentages in the precipitation samples resolved by EEM fluorescent spectroscopy combined with PARAFAC analysis.

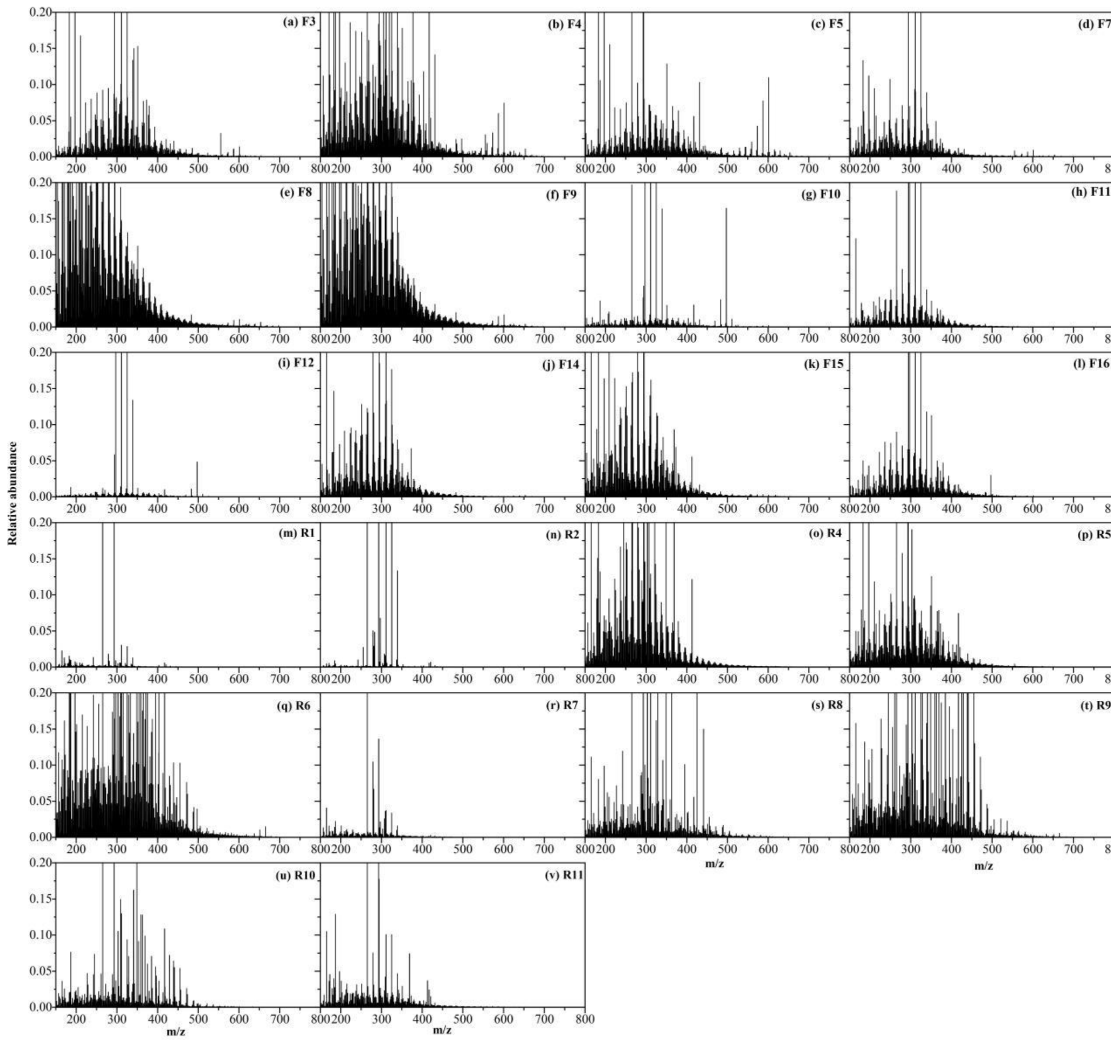

Figure S4. Reconstructed Fourier-transform ion cyclotron resonance mass spectra for 12 fog samples and 10 rainwater samples.

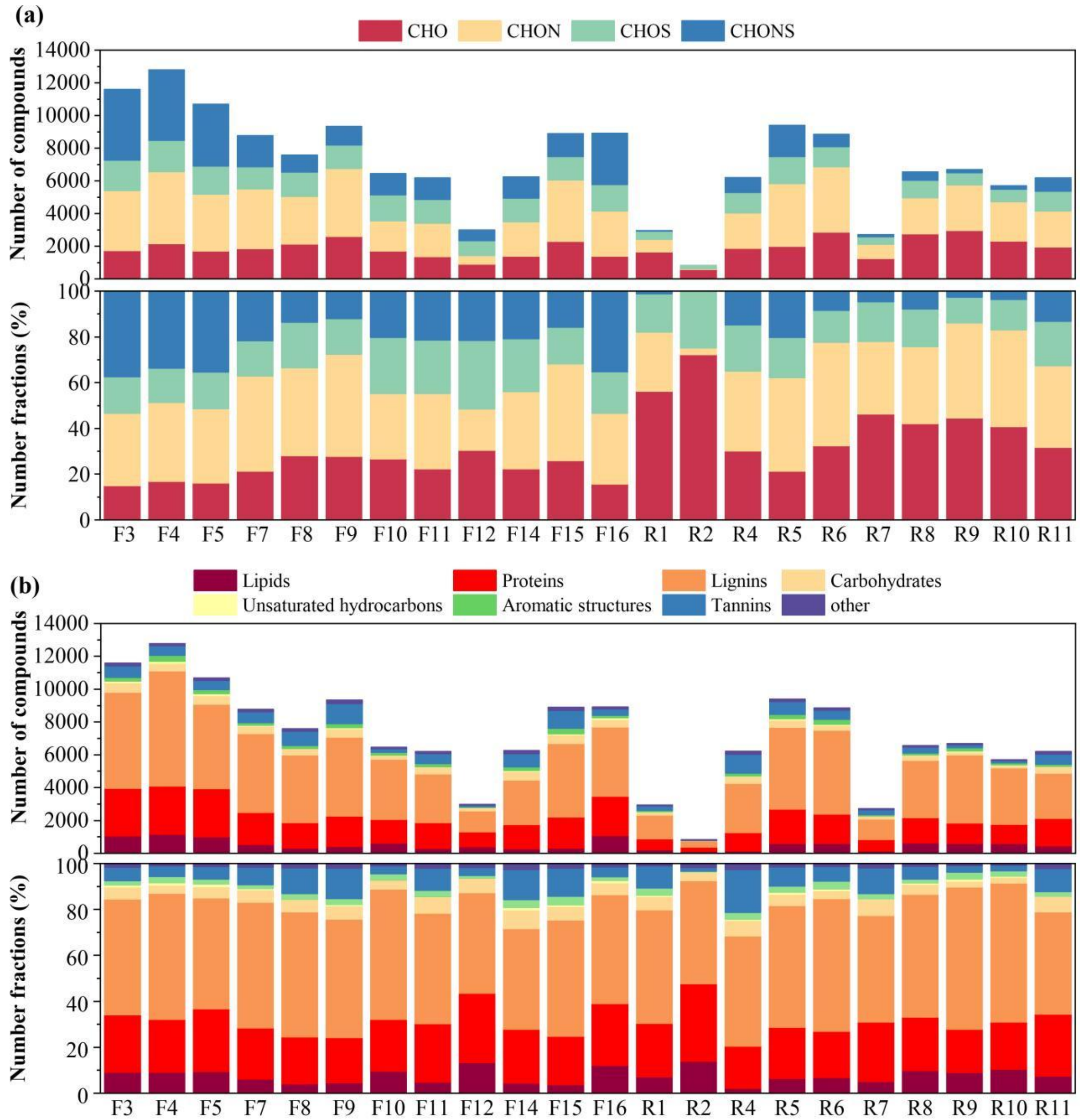

Figure S5. The total numbers and number fractions of (a) CHO, CHON, CHOS and CHONS, and (b) lipids, proteins, lignins, carbohydrates, unsaturated hydrocarbons, aromatic structures and tannins compounds in the precipitation samples.

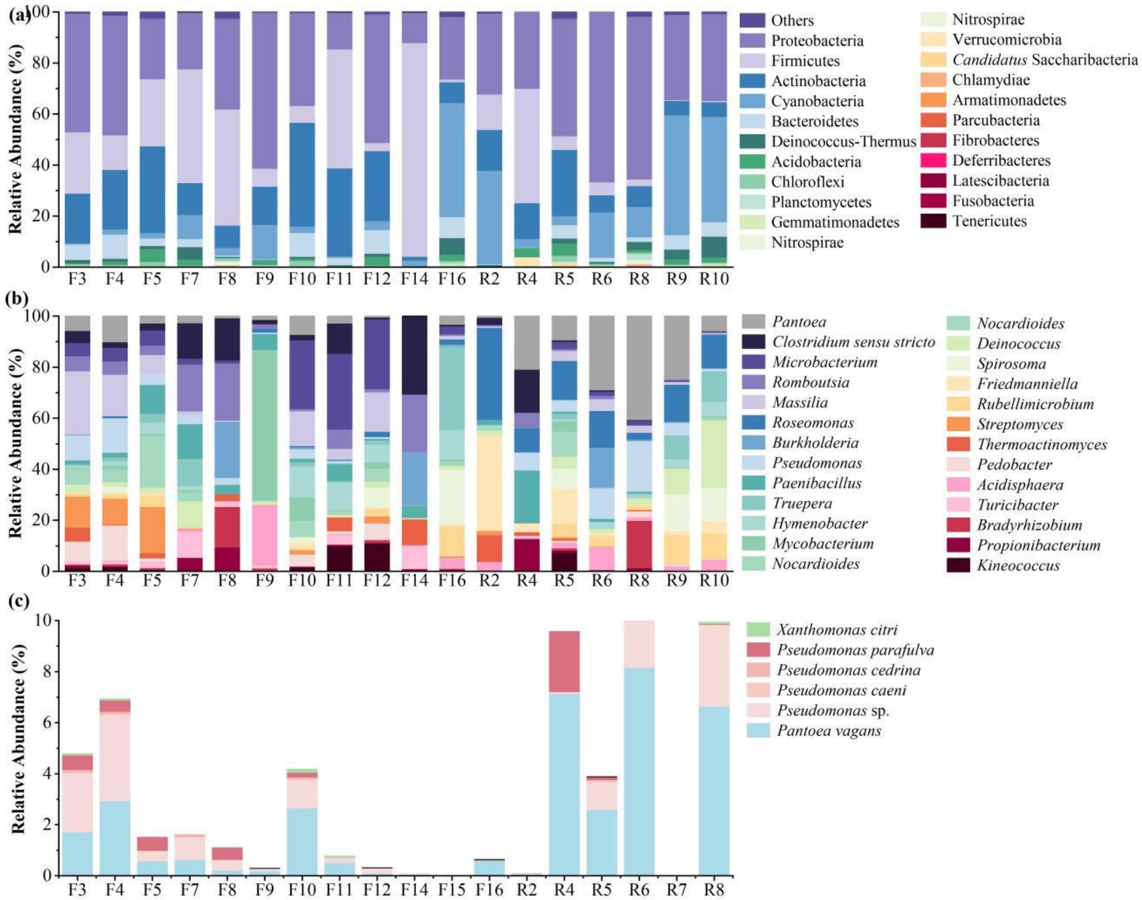

Figure S6. Relative abundances of the bacterial dominant (a) phyla, (b) genera and (c) the known INA species.

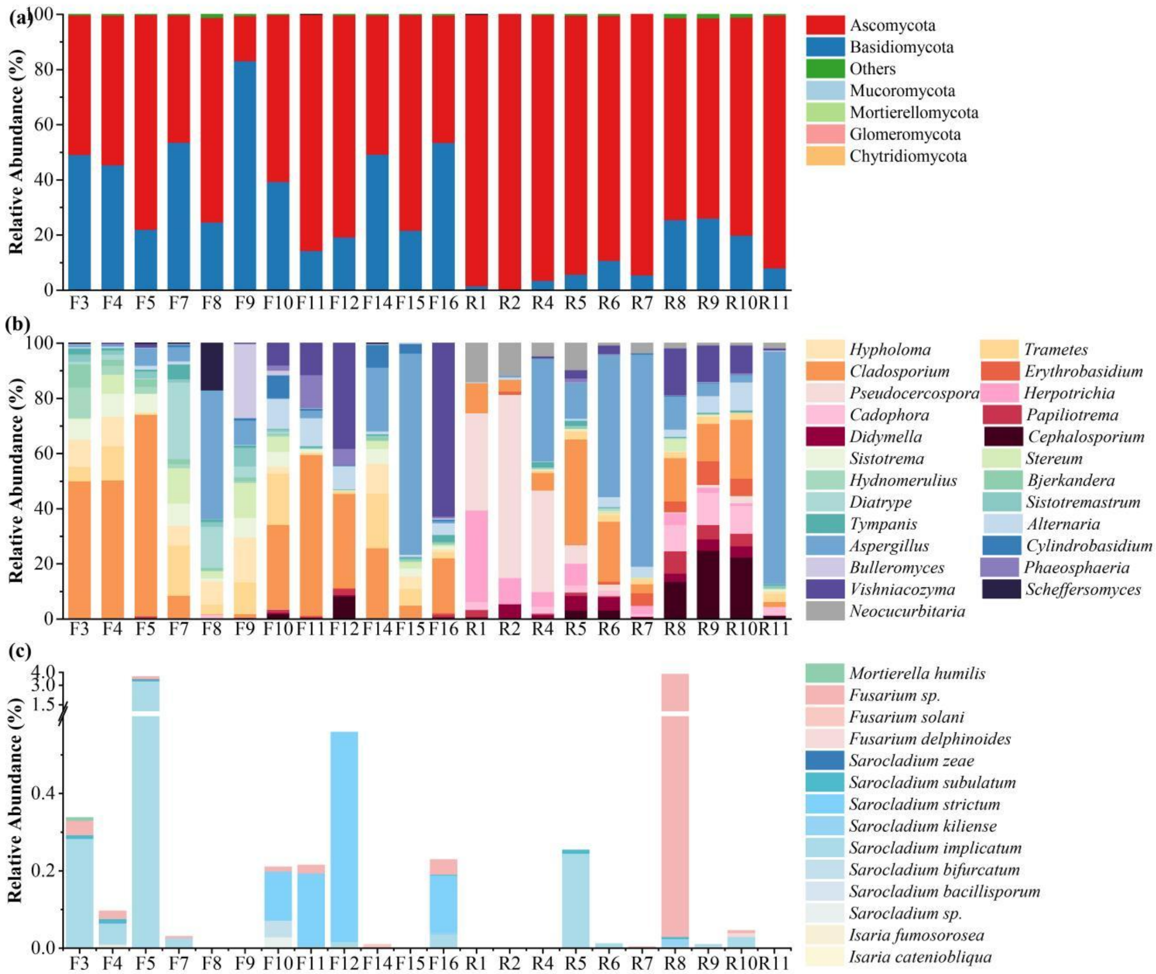

Figure S7. Relative abundances of the fungal dominant (a) phyla, (b) genera and (c) the known INA species.

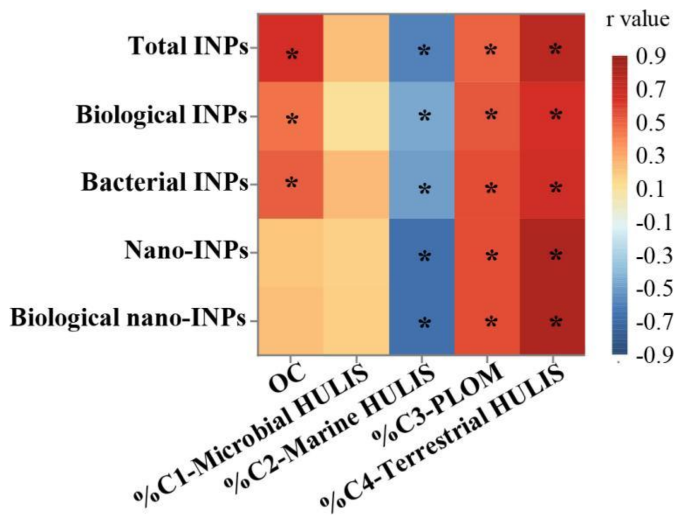

Figure S8. Spearman correlation heatmap of different types of INPs with (a) cations, (b) anions and (c) fluorescent components observed based on EEM data. The marker * represents the correlation that has significance at a p-value < 0.05.

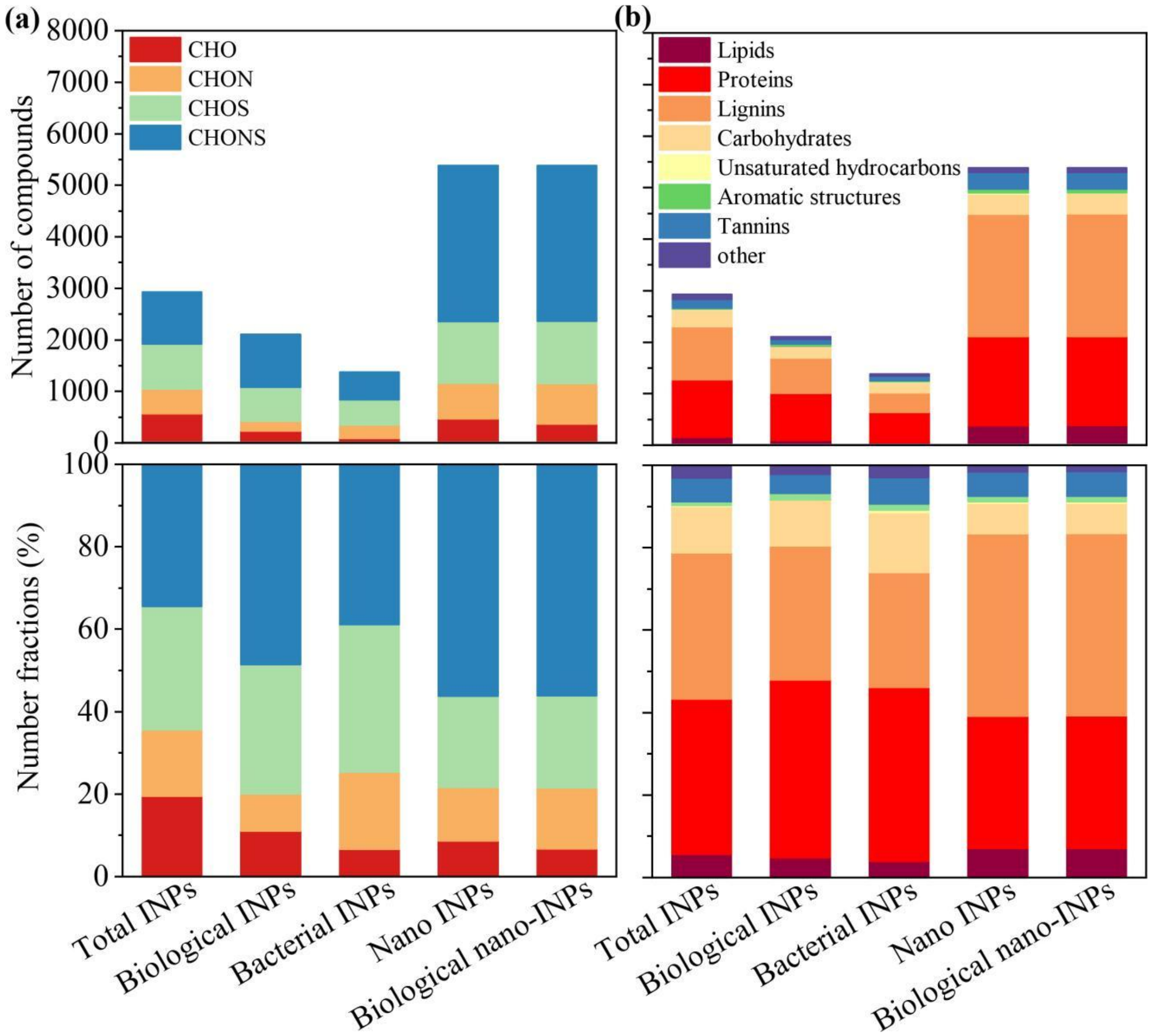

Figure S9. The total numbers and number fractions of (a) CHO, CHON, CHOS and CHONS, and (b) lipids, proteins, lignins, carbohydrates, unsaturated hydrocarbons, aromatic structures and tannins compounds related with different types of INPs based on Spearman's correlation analysis.

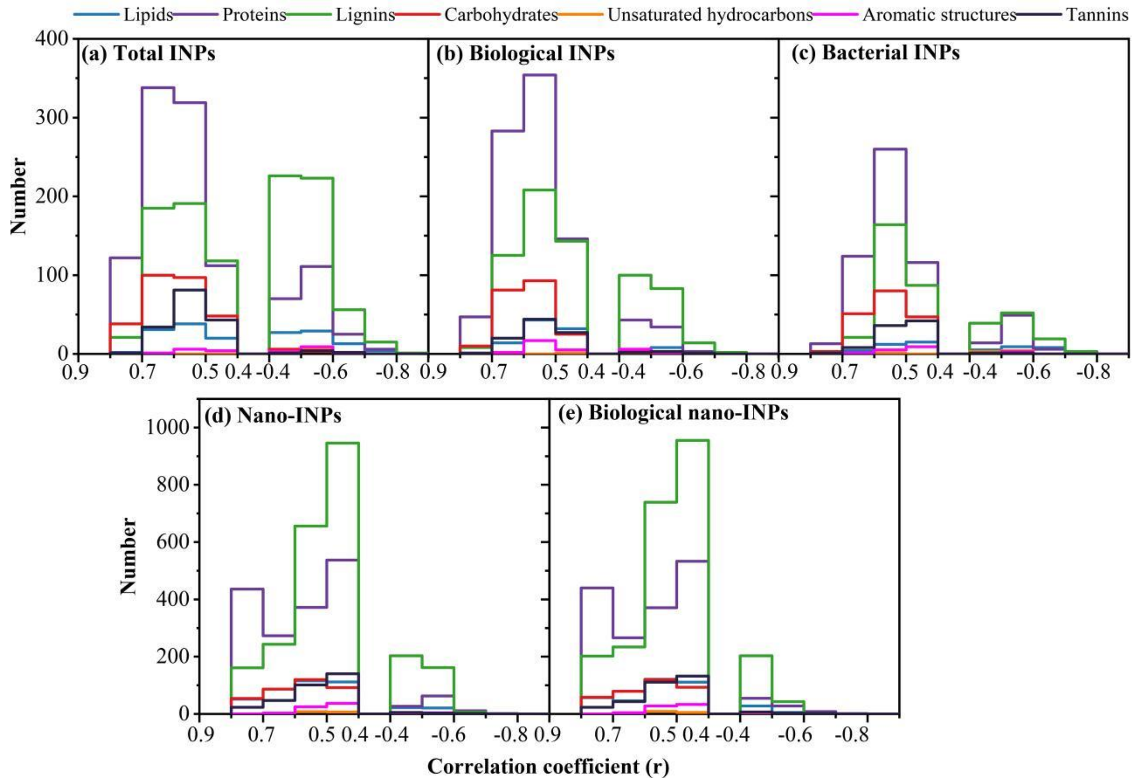

Figure S10. Correlation coefficient distribution of lipids, proteinaceous matter, lignins, carbohydrates, unsaturated hydrocarbons and tannins compounds with different types of INPs.

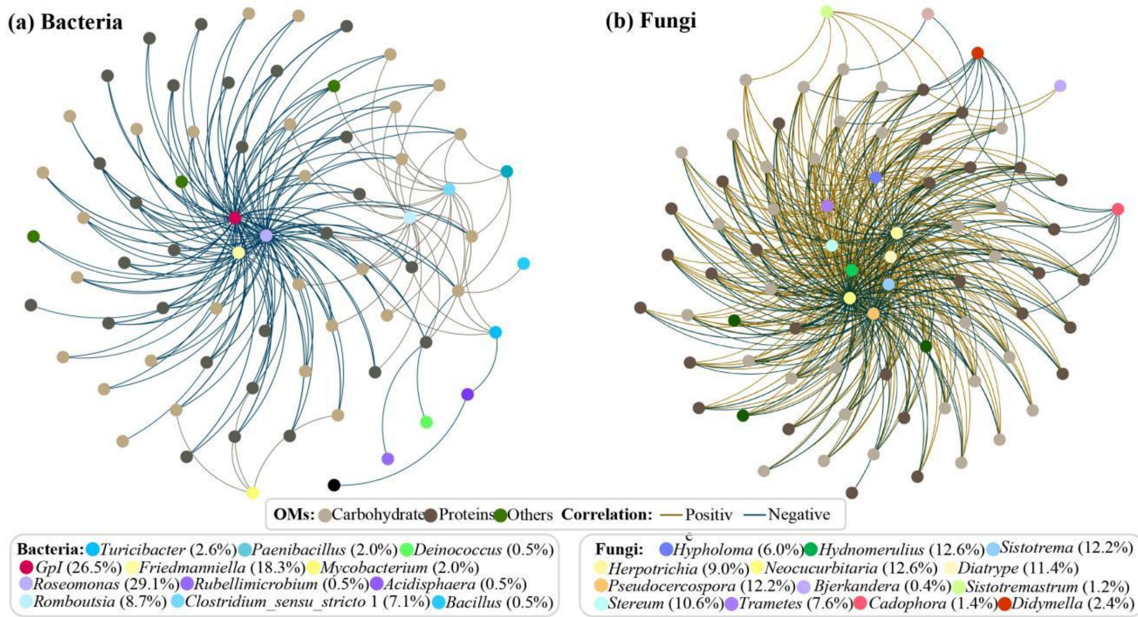

Figure S11. Co-occurrence analysis of the (a) bacterial and (b) fungal genera with OM molecules related to biological INPs (correlation coefficient > 0.7) based on Spearman's correlation analysis.

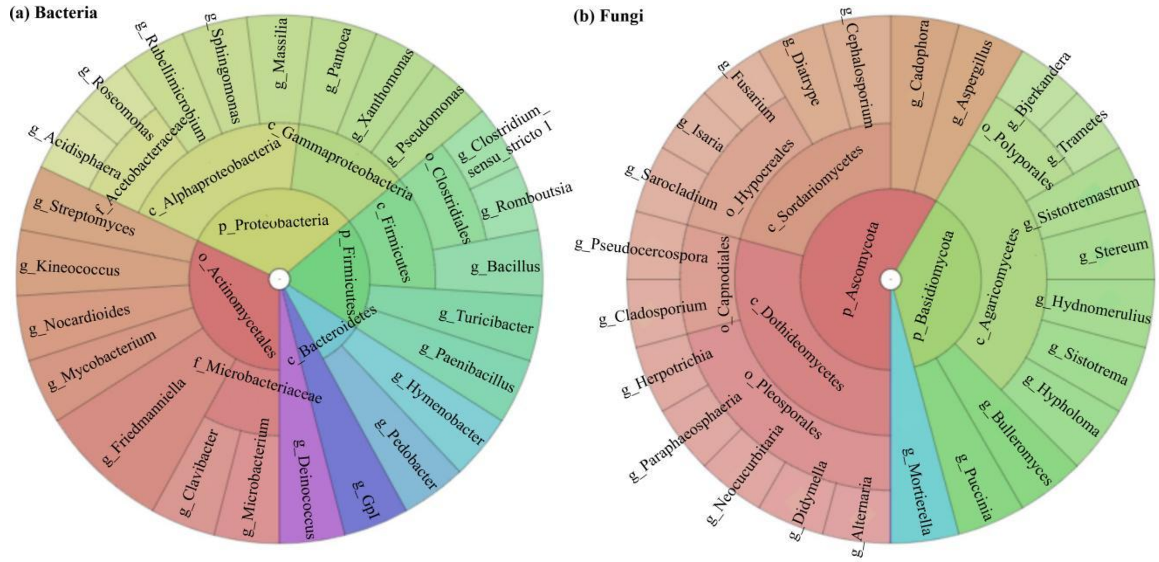

Figure S12. Krona diagram showing the taxonomic composition of the (a) bacterial and (b) fungal genera included in the co-occurrence network constructed between OM molecules associated with INPs (r values > 0.7) and microorganisms.

Table S1. The onset freezing temperature (T_0) and the temperature at which 50% of the droplets (T_{50}) of different types of INPs.

Sample	Sampling date (2019/##/## UTC+8:00)		INPs		Heat-resistant INPs		Lysozyme-resistant INPs		Nano-INPs		Heat-resistant nano-INPs	
ID	Start time	Stop time	T_0	T_{50}	T_0	T_{50}	T_0	T_{50}	T_0	T_{50}	T_0	T_{50}
F3	11/17 18:00	11/18 8:00	-5.8	-10.6	-10.2	-12.1	-5.5	-11.5	-6.2	-11.5	-10.6	-15.5
F4	11/18 8:30	11/18 16:30	-5.6	-7.7	-10.1	-16.8	-5.6	-8.7	-5.7	-11.8	-10.2	-23.9
F5	11/24 19:00	11/25 8:30	-7.6	-13.4	-11.2	-15.5	-7.6	-14.7	-10.3	-18.2	-11.7	-22.2
F6	11/25 8:30	11/25 20:00	-7.1	-14.4	-11.2	-18.2	-7.1	-14.9	-8.3	-19.7	-15.7	-23.1
F7	11/25 20:00	11/26 8:00	-7.0	-13.8	-11.8	-14.2	-7.9	-14.1	-8.1	-19.3	-12.7	-23.9
F8	11/26 8:30	11/26 19:30	-8.2	-15.1	-11.9	-17.2	-8.8	-15.6	-15.5	-21.7	-18.0	-24.4
F9	11/26 19:30	11/27 8:30	-6.0	-12.4	-11.9	-16.2	-6.2	-14.2	-7.5	-20.6	-12.8	-22.7
F10	11/27 8:30	11/27 19:30	-5.5	-7.5	-10.1	-14.6	-5.5	-8.0	-5.6	-8.7	-15.2	-21.5
F11	11/28 8:30	11/28 19:30	-6.0	-16.4	-12.7	-19.7	-6.0	-16.8	-8.7	-20.2	-19.2	-23.1
F12	11/28 20:00	11/29 8:30	-6.2	-12.1	-10.7	-19.3	-6.4	-13.9	-6.5	-12.8	-16.3	-22.0
F13	11/29 8:30	11/29 19:00	-5.9	-14.5	-10.4	-15.0	-6.4	-14.9	-8.6	-19.5	-16.6	-22.7
F14	11/30 8:30	11/30 19:30	-10.2	-14.0	-10.9	-14.8	-10.2	-14.7	-10.2	-19.6	-18.3	-23.9
F15	11/30 19:30	12/01 9:30	-11.2	-20.1	-13.5	-22.1	-11.4	-19.2	-16.1	-24.4	-18.0	-25.8
F16	12/01 19:30	12/02 9:00	-6.0	-13.0	-10.3	-15.4	-6.0	-14.3	-6.5	-18.2	-16.4	-20.1
Fog			**-7.0**	**-13.2**	**-11.2**	**-16.5**	**-7.2**	**-14.0**	**-8.8**	**-17.6**	**-15.1**	**-22.5**
R1	11/12 2:00	11/12 7:30	-14.1	-23.9	-19.3	-24.3	-12.8	-23.6	-11.0	-25.1	-21.4	-26.4
R2	11/12 7:30	11/12 11:00	-14.8	-22.8	-15.6	-23.0	-14.3	-23.0	-18.2	-24.8	-18.2	-25.1
R4	11/13 0:00	11/13 9:00	-9.8	-20.6	-11.5	-20.2	-11.8	-21.2	-15.5	-23.9	-17.2	-24.4
R5	11/17 18:00	11/18 7:00	-10.0	-20.0	-12.0	-21.2	-11.0	-20.6	-10.1	-21.3	-11.0	-23.6
R6	11/18 9:00	11/18 13:00	-9.6	-20.0	-12.5	-23.2	-10.9	-20.8	-12.9	-21.5	-16.9	-24.1
R7	11/26 9:00	11/26 10:00	-11.7	-19.8	-14.0	-24.1	-13.2	-20.1	-14.2	-20.4	-18.2	-25.7
R8	11/27 2:00	11/27 8:00	-10.4	-20.4	-11.4	-21.9	-14.0	-21.9	-12.3	-22.1	-14.7	-23.9
R9	11/27 10:00	11/27 12:00	-11.8	-21.9	-13.0	-23.5	-12.7	-23.3	-6.2	-23.2	-15.5	-23.5
R10	11/27 14:00	11/27 18:00	-10.0	-21.6	-12.4	-22.7	-10.8	-22.7	-11.3	-22.5	-16.9	-23.5
R11	12/01 4:00	12/01 9:30	-12.4	-21.2	-14.4	-21.4	-13.5	-22.8	-17.3	-22.7	-17.9	-24.5
Rain			**-11.5**	**-21.2**	**-13.6**	**-22.6**	**-12.5**	**-22.0**	**-12.9**	**-22.8**	**-16.8**	**-24.5**

Table S2. Spearman correlation between different types of INPs and the number of different categories of organic molecular formulas assigned according to the elemental compositions.

Types of INPs	CHO	CHON	CHOS	CHONS	Total formulas
Total INPs	-0.04	0.43	0.78**	0.77**	0.63**
Biological INPs	-0.01	0.41	0.69**	0.62**	0.52*
Bacterial INPs	0.17	0.29	0.58**	0.46*	0.52*
Nano-INPs	-0.33	0.19	0.59**	0.64**	0.37
Biological nano-INPs	-0.37	0.14	0.59**	0.64**	0.33

Note: * $p < 0.05$, ** $p < 0.01$

Table S3. The information of OM molecules exhibiting negative correlations with INPs. The possible naming of compounds was provided.

m/z	OM formula	H/C	O/C	R^2	Possible molecule
186	$C_9H_{17}NO_3$	1.89	0.33	-0.56	Ac-Ile-OMe
230	$C_{11}H_{21}NO_4$	1.91	0.36	-0.51	Boc-D-Ile-OH
244	$C_{12}H_{23}NO_4$	1.92	0.33	-0.54	Boc-D-Leu-OMe
202	$C_9H_{17}NO_4$	1.89	0.44	-0.56	Ac-Ser(tBu)-OH Boc-N-methyl-L-alanine
284	$C_{12}H_{19}N_3O_5$	1.58	0.42	-0.62	H-Gly-Pro-Hyp-OH
257	$C_{12}H_{22}N_2O_4$	1.83	0.33	-0.63	Boc-Pro-NMe(OMe)
286	$C1_2H_{21}N_3O_5$	1.75	0.42	-0.61	Ac-Ala-Ala-Ala-OMe
170	$C_9H_{17}NO_2$	1.89	0.22	-0.55	H-D-Pro-OtBu
442	$C_{21}H_{37}N_3O_7$	1.76	0.33	-0.47	Boc-Lys(Boc)-Pro-OH
245	$C_{10}H_{18}N_2O_5$	1.80	0.50	-0.59	Boc-Gln-OH
287	$C_{13}H_{24}N_2O_5$	1.85	0.38	-0.48	Boc-Leu-Gly-OH
411	$C_{23}H_{28}N_2O_5$	1.22	0.22	-0.47	Z-Phe-Leu
425	$C_{23}H_{26}N_2O_6$	1.13	0.26	-0.61	Boc-D-Gln(Xan)-OH