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Measurement report: Molecular characteristics of cloud water in southern China and insights into aqueous-phase processes from Fourier transform ion cyclotron resonance mass spectrometry

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Text S1. Meteorological conditions and water-soluble ions in cloud water

During the investigated period (May 11-12), air masses from the South China Sea and Southeast Asia and across Hainan Island and the boundary between Guangdong and Guangxi Provinces were dominant, according to the back-trajectory analysis (Fig. S1). As presented in Fig. S2, there is no obvious diurnal variation of temperature (15.2 -15.9 ℃). As expected, the relative humidity was stable, with a constant value of 100%. The wind with speeds ranged from 3.3 to 7.5 m s^{-1} and primarily came from the southwest. The $PM_{2.5}$ concentration varied from 2.0 to 4.3 μg m^{-3}, much lower than that in most urban areas in southern China. The NO_x, O_3, and SO_2 concentrations ranged from 2.2 to 7.7 μg m^{-3}, 60.6 to 101.0 μg m^{-3}, and 0.2 to 0.8 μg m^{-3}, respectively. The NO_x and O_3 concentrations showed a clear diurnal variation. Both of them were relatively stable in the daytime (approximately 2-4 μg m^{-3} and 85-100 μg m^{-3} for NO_x and O_3, respectively). However, NO_x reached a peak of 8 μg m^{-3} at approximately 01:00 LT, May 12, and then decreased, whereas O_3 dropped from approximately 100 μg m^{-3} at about 21:00, May 11 to 60 μg m^{-3} in the next morning (Fig. S2).

The concentrations of inorganic ions in cloud water are listed in Table S1. Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, and SO_4^{2-} were detected. SO_4^{2-}, NO_3^-, and NH_4^+ were dominant, with average concentrations of 3.53, 2.88, and 1.11 mg L^{-1}, respectively; together, they contributed approximately 90% of the total water-soluble inorganic ions. These concentrations are comparable to those observed in cloud water collected at Mt. Heng (Sun et al., 2010) and Mt. Lu (Sun et al., 2016) in southern China, but much lower than those observed at Mt. Tai in northern China (Guo et al., 2012).

References

Guo, J., Wang, Y., Shen, X., Wang, Z., Lee, T., Wang, X., Li, P., Sun, M., Collett, J. L., Wang, W., and Wang, T.: Characterization of cloud water chemistry at Mount Tai, China: Seasonal variation, anthropogenic impact, and cloud processing, Atmos. Environ., 60, 467-476, https://doi.org/10.1016/j.atmosenv.2012.07.016, 2012.

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Sun, X., Wang, Y., Li, H., Yang, X., Sun, L., Wang, X., Wang, T., and Wang, W.: Organic acids in cloud water and rainwater at a mountain site in acid rain areas of South China, Environ. Sci. Pollut. Res. Int., 23, 9529-9539, https://doi.org/10.1007/s11356-016-6038-1, 2016.

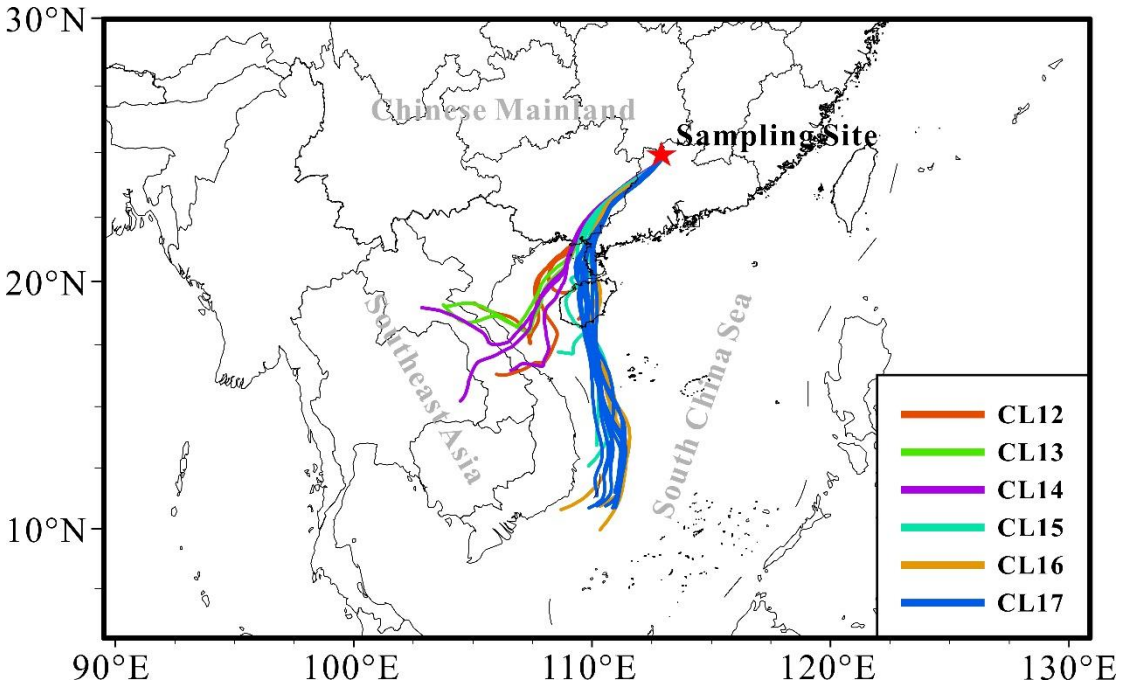

Figure S1. The location of the sampling site and 72 h back trajectories displayed by the Hybrid Single-Particle Lagrangian Integrated Trajectory model for every hours during sampling time (May 11 10:00 – May 12 08:00 LT).

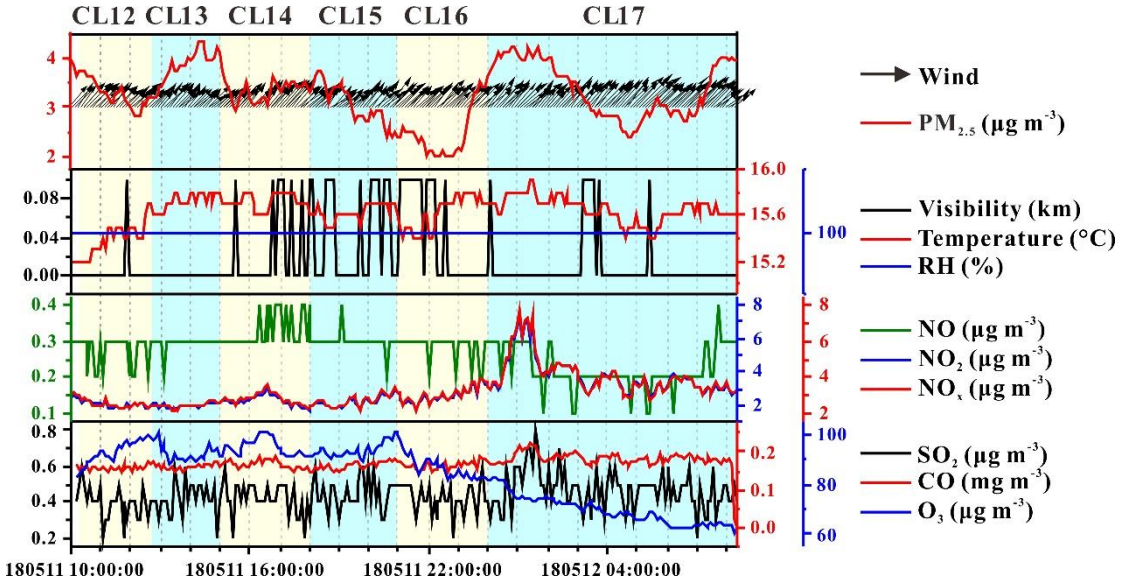

Figure S2. The time series of the meteorological data including visibility, temperature, relative humidity, concentration of some contaminant in the atmosphere (NO, NO_2, NO_x, SO_2, CO, and O_3). The data was obtained from Guangdong Environmental Monitoring Center. The alternating color lumps in the figure represent the sampling interval.

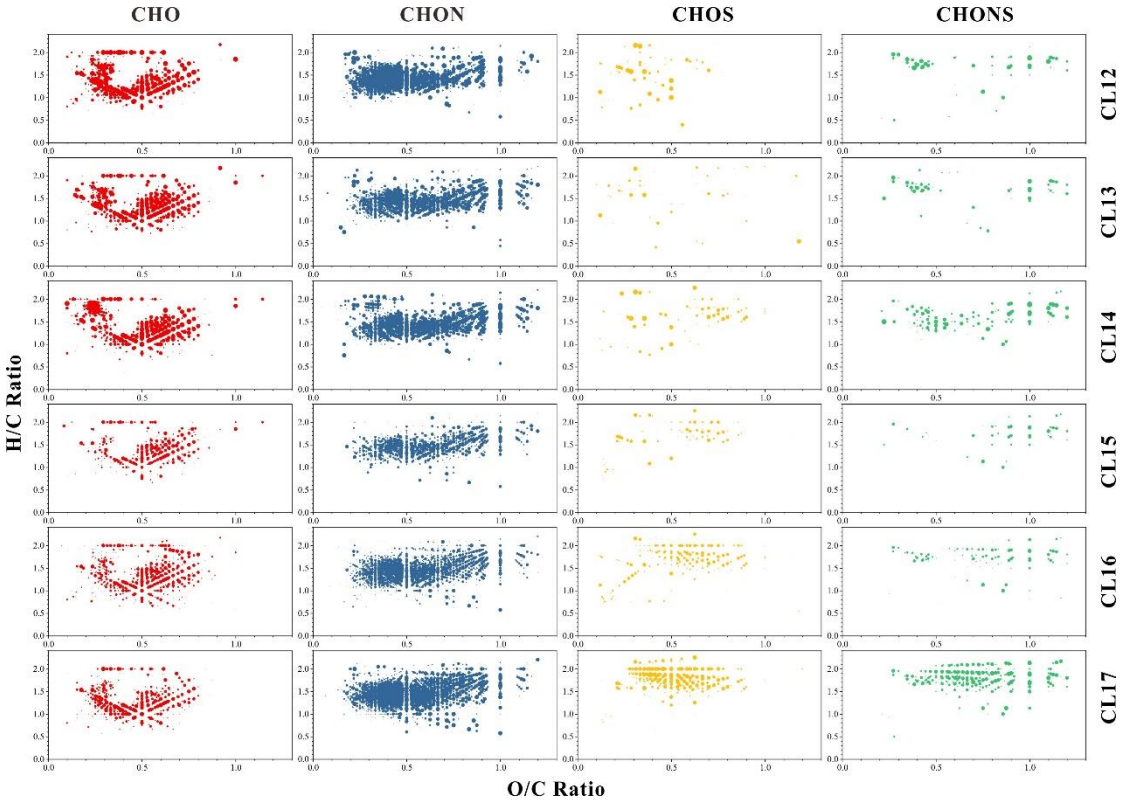

Figure S3. Van Krevelen diagrams as a function of four groups (CHO, CHNO, CHOS, and CHNOS) for the cloud water samples. The larger point in the diagram represents the higher relative abundance of the formula.

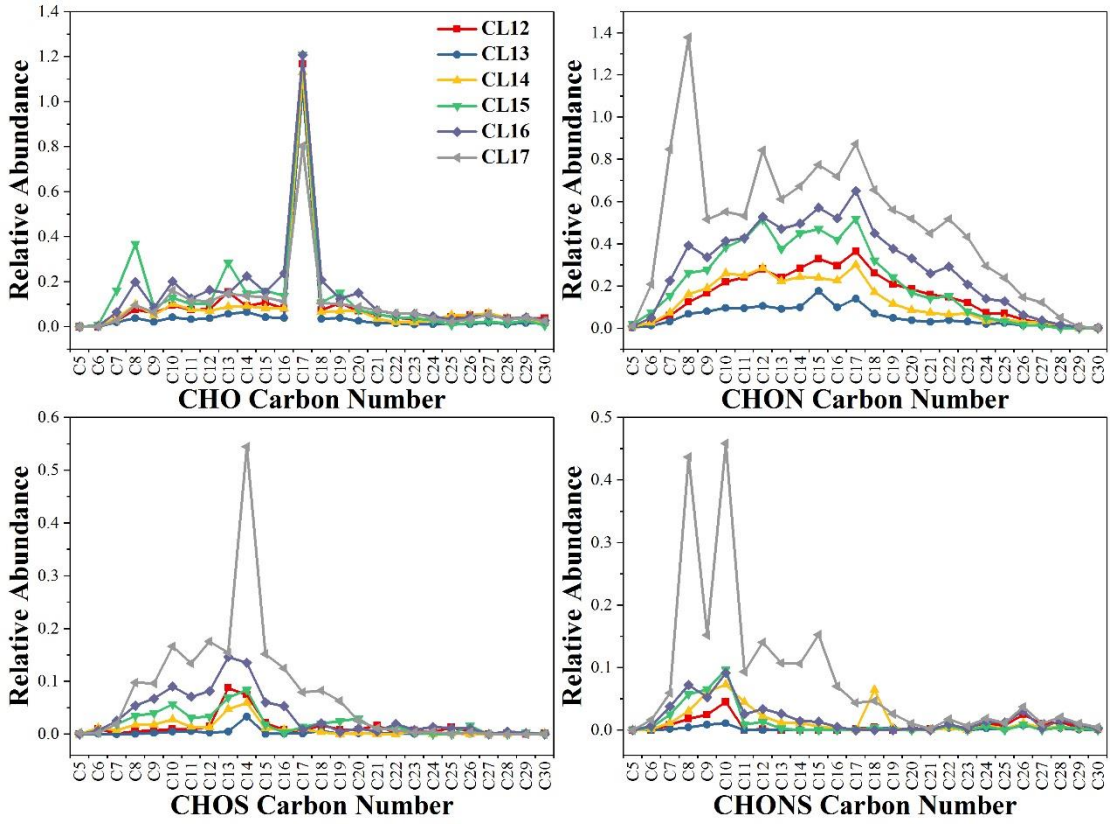

Figure S4. The relative abundances with respect to the carbon atom number within four groups (CHO, CHON, CHOS, and CHONS) of the molecular formulas in the cloud water samples.

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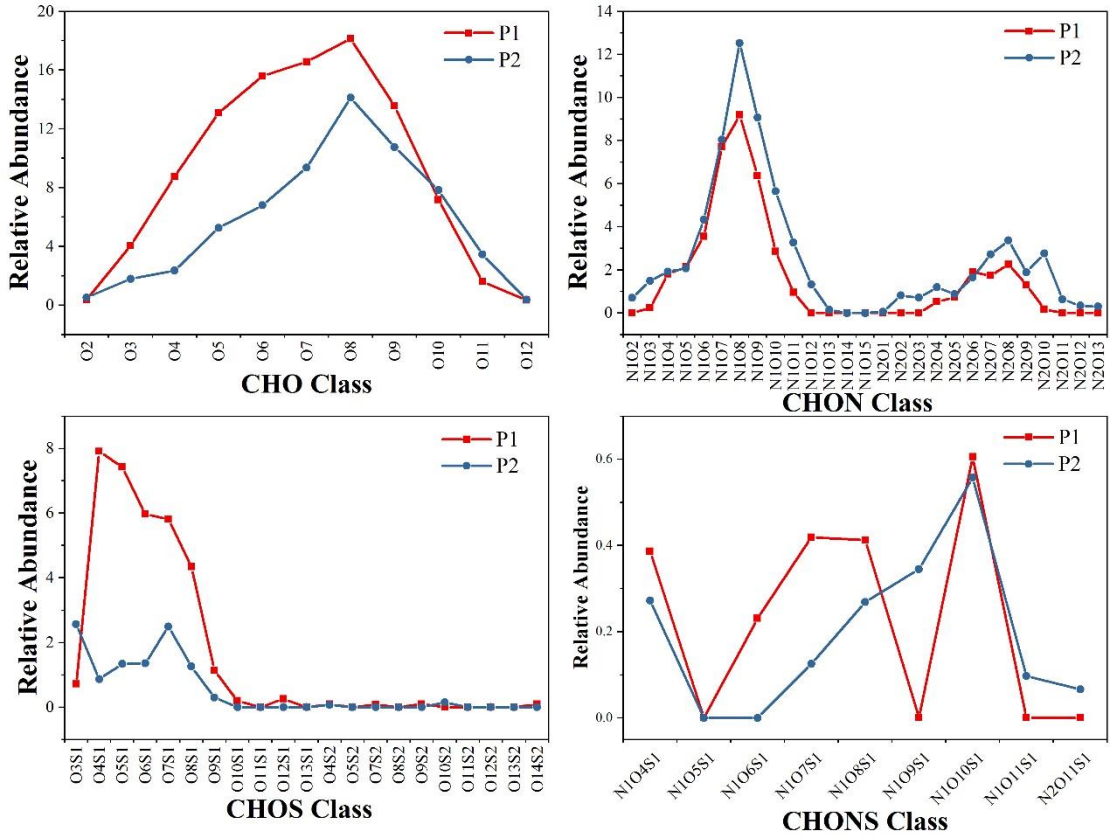

Figure S5. Relative abundance of the categories of CHO, CHON, CHOS and CHONS formulas according to the characteristic atom groups in the molecular formulas in two $PM_{2.5}$ samples (P1 and P2).

Table S1. The concentration of water-soluble inorganic ions in cloud water. The unit of the data is mg L^{-1}.

Sample ID	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	$C_2O_4^{2-}$
CL12	0.17	0.61	0.04	0.04	0.01	0.34	2.21	2.66	0.19
CL13	0.27	0.80	0.09	0.03	0.02	0.27	2.30	2.74	0.30
CL14	0.23	0.91	0.05	0.03	N.D.	0.27	2.30	2.74	0.47
CL15	0.29	1.23	0.08	0.05	0.04	0.37	3.11	4.05	0.39
CL16	0.25	1.46	0.09	0.05	0.03	0.46	3.44	4.23	0.06
CL17	0.37	1.69	0.10	0.06	0.03	0.67	3.93	4.74	N.D.

N.D.: Not detected.

Table S2. The number fraction and relative abundance fraction of four groups (CHO, CHON, CHOS, and CHONS) in each cloud water sample.

Sample ID	Number Fraction				Relative Abundance Fraction			
	CHO	CHON	CHOS	CHONS	CHO	CHON	CHOS	CHONS
CL12	26.6%	65.2%	4.5%	3.7%	37.6%	54.9%	4.7%	2.7%
CL13	28.3%	64.4%	3.6%	3.7%	52.1%	43.6%	2.3%	2.0%
CL14	27.1%	60.2%	4.9%	7.8%	39.3%	50.5%	4.1%	6.1%
CL15	23.5%	66.2%	6.4%	3.9%	35.3%	56.2%	5.3%	3.1%
CL16	21.7%	65.4%	8.1%	4.8%	30.0%	58.9%	7.3%	3.9%
CL17	16.3%	65.1%	9.4%	9.3%	13.8%	65.3%	10.2%	10.8%

Table S3. The number fraction and relative abundance fraction of four groups (CHO, CHON, CHOS, and CHONS) in two $PM_{2.5}$ samples.

Sample ID	Number Fraction				Relative Abundance Fraction			
	CHO	CHON	CHOS	CHONS	CHO	CHON	CHOS	CHONS
P1	49.8%	31.8%	17.2%	1.2%	55.5%	24.3%	19.1%	1.2%
P2	39.9%	51.0%	7.5%	1.6%	44.0%	47.3%	7.5%	1.2%

Table S4. The values of average O/C, H/C, DBE, number of carbon (#C) and OS_C and relative-abundance-weighted values of that for all formulas, CHO, CHON, CHOS and CHONS groups in cloud water.

All										
Sample ID	O/C	H/C	DBE	#C	OS_C	O/Cw	H/Cw	DBEw	#Cw	OS_{Cw}
CL12	0.46	1.44	6.16	16.92	-0.88	0.45	1.47	5.70	16.16	-0.87
CL13	0.52	1.45	5.96	16.31	-0.76	0.45	1.48	5.37	15.82	-0.81
CL14	0.53	1.46	5.61	15.91	-0.82	0.51	1.48	5.25	15.22	-0.79
CL15	0.56	1.47	5.61	15.20	-0.76	0.56	1.49	5.10	14.38	-0.72
CL16	0.52	1.46	5.92	16.47	-0.83	0.54	1.48	5.41	15.28	-0.79
CL17	0.51	1.49	5.86	16.80	-0.92	0.55	1.44	5.44	14.58	-0.91
CHO										
Sample ID	O/C	H/C	DBE	#C	OS_C	O/Cw	H/Cw	DBEw	#Cw	OS_{Cw}
CL12	0.41	1.38	6.37	18.18	-0.57	0.36	1.45	5.59	16.98	-0.72
CL13	0.46	1.38	6.30	17.67	-0.46	0.34	1.47	5.33	16.69	-0.80
CL14	0.44	1.41	5.82	17.98	-0.53	0.38	1.46	5.29	16.78	-0.71
CL15	0.50	1.39	5.79	15.93	-0.39	0.46	1.47	4.96	14.94	-0.54
CL16	0.47	1.36	6.35	17.53	-0.41	0.45	1.46	5.27	16.11	-0.56
CL17	0.43	1.31	6.85	17.50	-0.46	0.39	1.36	6.12	16.44	-0.58
CHON										
Sample ID	O/C	H/C	DBE	#C	OS_C	O/Cw	H/Cw	DBEw	#Cw	OS_{Cw}
CL12	0.48	1.45	6.24	16.46	-0.98	0.51	1.45	6.00	15.65	-0.92
CL13	0.54	1.45	5.99	15.66	-0.86	0.59	1.48	5.50	14.75	-0.78
CL14	0.55	1.46	5.84	15.64	-0.87	0.60	1.46	5.50	14.46	-0.76
CL15	0.57	1.47	5.81	15.25	-0.84	0.62	1.48	5.44	14.27	-0.76
CL16	0.53	1.46	6.24	16.60	-0.90	0.57	1.44	5.88	15.23	-0.83
CL17	0.50	1.46	6.41	17.31	-0.94	0.56	1.37	6.09	14.77	-0.87
CHOS										
Sample ID	O/C	H/C	DBE	#C	OS_C	O/Cw	H/Cw	DBEw	#Cw	OS_{Cw}
CL12	0.39	1.56	4.92	16.20	-1.22	0.36	1.66	3.87	15.11	-1.39

CL13	0.50	1.58	4.75	16.16	-1.03	0.45	1.56	4.58	15.15	-1.11
CL14	0.59	1.58	3.81	12.35	-0.99	0.50	1.66	3.19	12.60	-1.21
CL15	0.50	1.61	4.35	13.93	-1.11	0.47	1.68	3.72	13.77	-1.23
CL16	0.52	1.65	3.64	13.74	-1.09	0.51	1.71	3.21	13.27	-1.18
CL17	0.50	1.74	2.89	14.10	-1.21	0.48	1.75	2.72	13.43	-1.27
CHONS										
Sample ID	O/C	H/C	DBE	#C	OS_C	O/Cw	H/Cw	DBEw	#Cw	OS_{Cw}
CL12	0.68	1.63	4.63	17.03	-1.14	0.69	1.67	4.29	16.96	-1.19
CL13	0.67	1.71	4.12	17.42	-1.21	0.67	1.71	4.13	17.28	-1.21
CL14	0.67	1.58	4.22	13.10	-1.31	0.65	1.60	4.25	13.22	-1.33
CL15	0.84	1.72	3.18	11.98	-1.15	0.88	1.72	3.01	10.92	-1.14
CL16	0.74	1.71	3.48	14.47	-1.17	0.78	1.68	3.42	13.33	-1.16
CL17	0.70	1.77	3.29	14.75	-1.27	0.78	1.70	3.19	12.09	-1.21

Table S5. The relative-abundance-weighted average values of O/C, H/C, DBE, OS_C and for all formulas, CHO, CHON, CHOS and CHONS formulas in two $PM_{2.5}$ samples (P1 and P2).

Group	Sample ID	O/C_w	H/C_w	DBE_w	OS_{Cw}
All	P1	0.45	1.53	5.04	-0.84
	P2	0.56	1.40	4.74	-0.61
CHO	P1	0.40	1.49	5.48	-0.69
	P2	0.53	1.45	5.03	-0.40
CHON	P1	0.53	1.51	5.32	-0.89
	P2	0.63	1.49	5.04	-0.75
CHOS	P1	0.46	1.65	3.49	-1.19
	P2	0.61	1.39	4.73	-0.83
CHONS	P1	0.66	1.69	3.22	-1.40
	P2	0.86	1.74	2.98	-1.17

Table S6. The fraction in relative abundance of aliphatic/olefinic and (condensed) aromatic structures in six cloud water samples.

CHO						
Sample ID	CL12	CL13	CL14	CL15	CL16	CL17
Aliphatic/Olefinic	95.8%	98.3%	97.1%	97.2%	94.9%	91.1%
(Condensed) Aromatic	4.2%	1.7%	2.9%	2.8%	5.1%	8.9%
CHON						
Sample ID	CL12	CL13	CL14	CL15	CL16	CL17
Aliphatic/Olefinic	96.1%	97.5%	97.2%	97.1%	92.5%	79.2%
(Condensed) Aromatic	3.9%	2.5%	2.8%	2.9%	7.5%	20.8%
CHOS						
Sample ID	CL12	CL13	CL14	CL15	CL16	CL17
Aliphatic/Olefinic	93.9%	95.3%	96.3%	93.5%	95.7%	98.8%
(Condensed) Aromatic	6.1%	4.7%	3.7%	6.5%	4.3%	1.2%
CHONS						
Sample ID	CL12	CL13	CL14	CL15	CL16	CL17
Aliphatic/Olefinic	95.7%	98.2%	100.0%	100.0%	98.6%	99.6%
(Condensed) Aromatic	4.3%	1.8%	0.0%	0.0%	1.4%	0.4%