

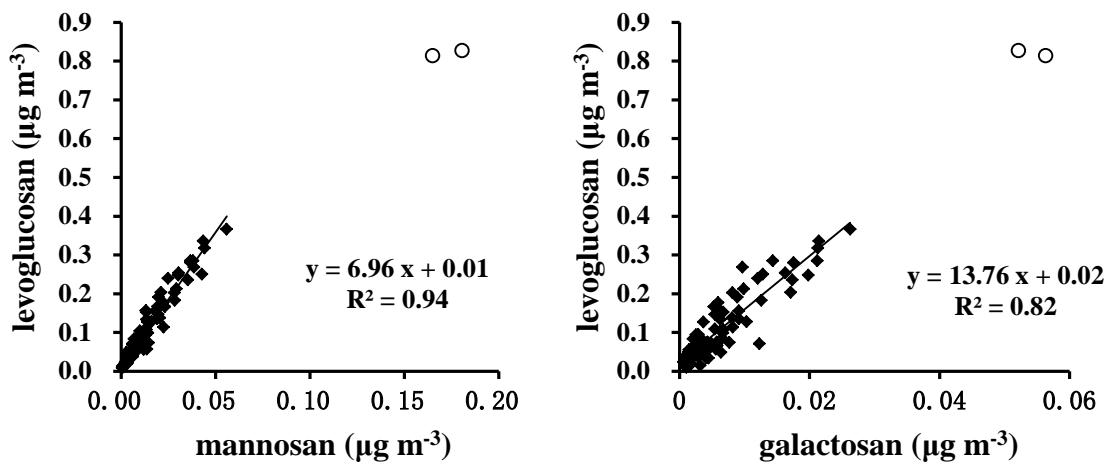


*Supplement of*

## **Sources of humic-like substances in the Pearl River Delta, China: positive matrix factorization analysis of PM<sub>2.5</sub> major components and source markers**

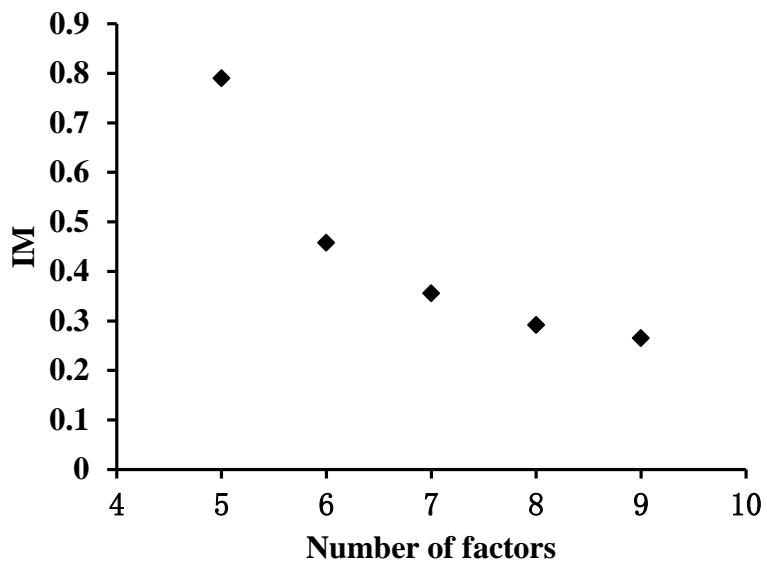
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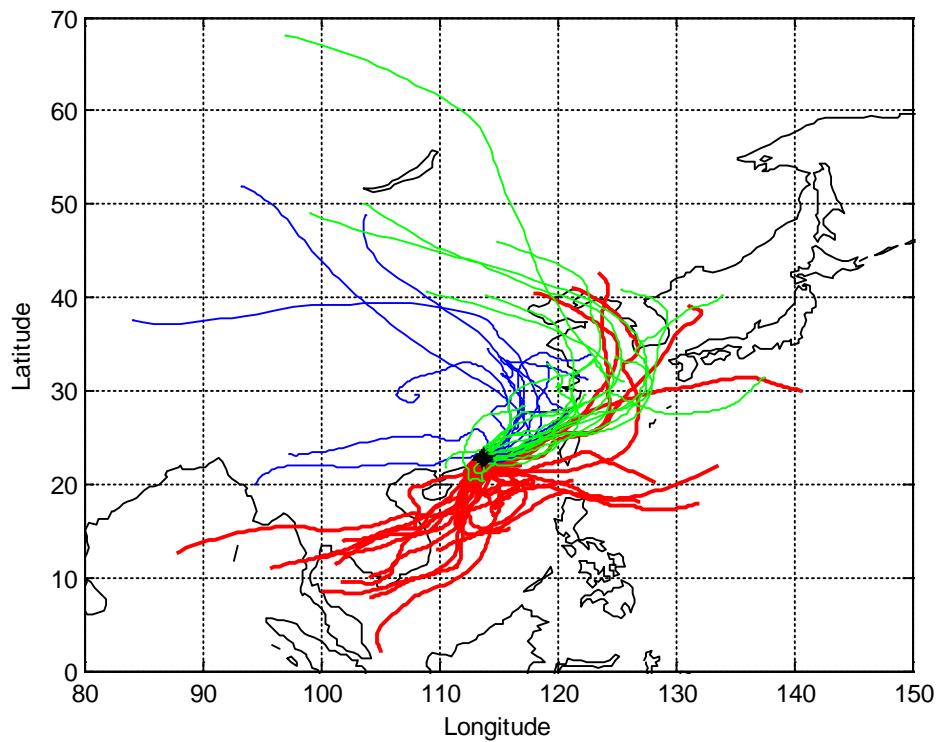


**Fig. S1** Relationship between levoglucosan and mannosan, galactosan.

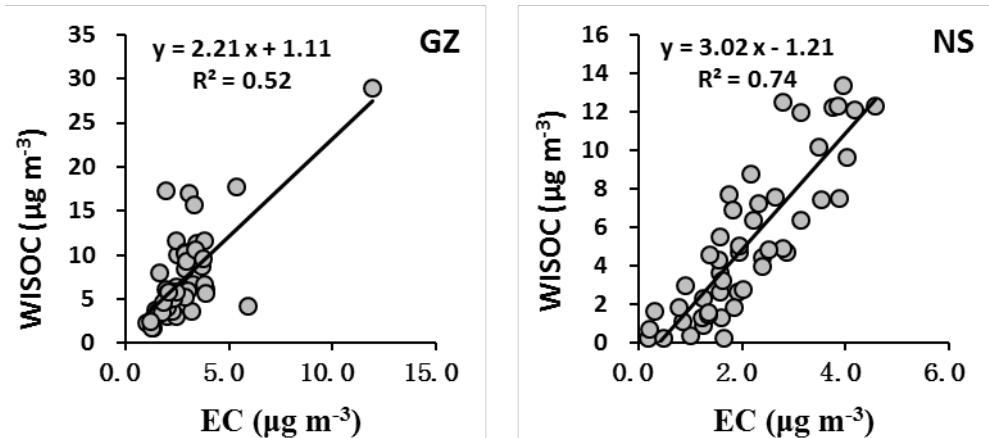
The open circles represent the points which were not input into PMF (GZ Jan 26, NS Jan 26, because of extremely high levoglucosan)



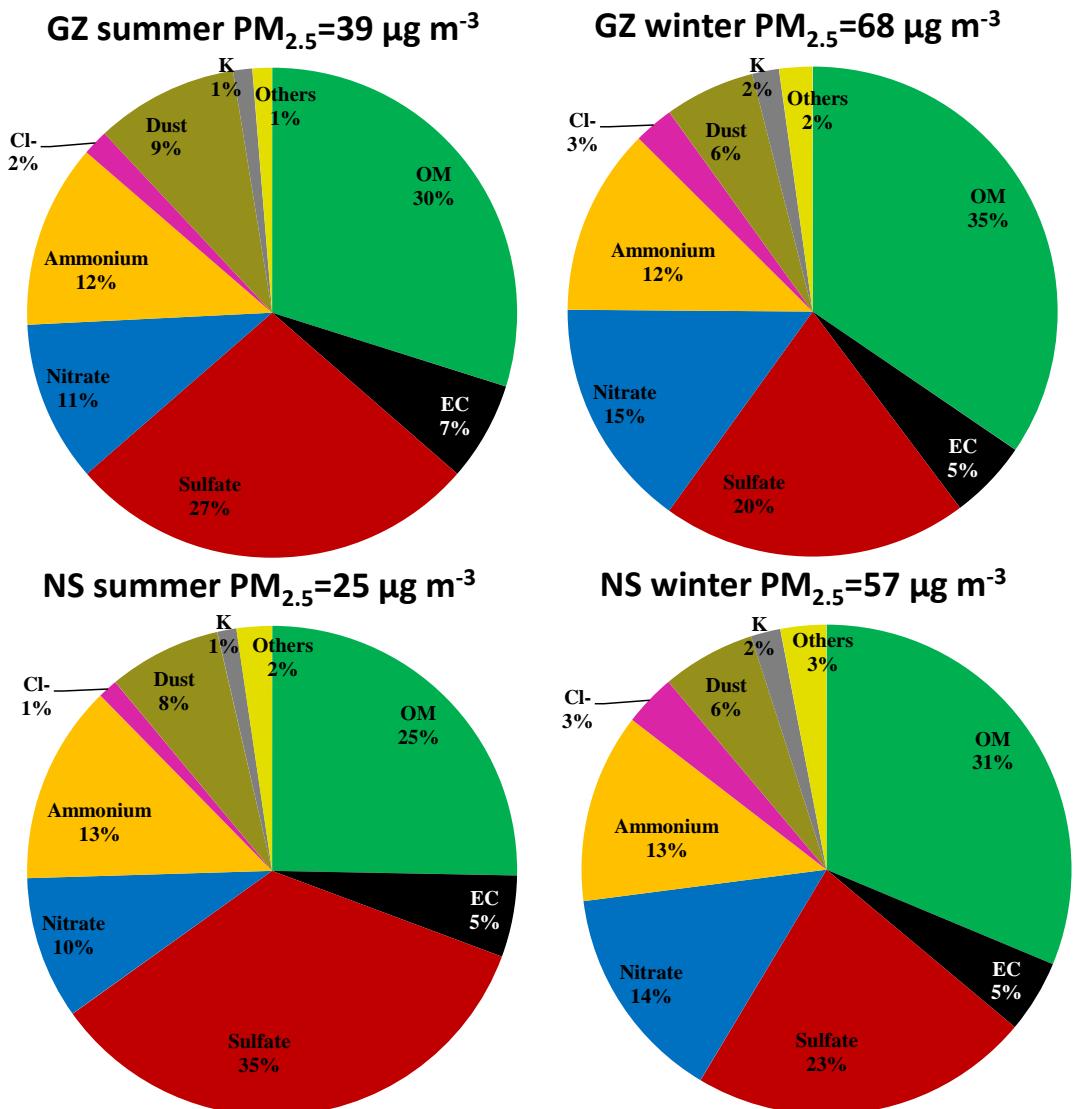
**Fig. S2** IM (maximum Individual column Mean) values of different factor numbers



**Fig. S3** Back trajectories of all sampling days in 2009, with lines in red, green, and blue representing air mass back trajectories of marine, transitional, and continental origins, respectively.



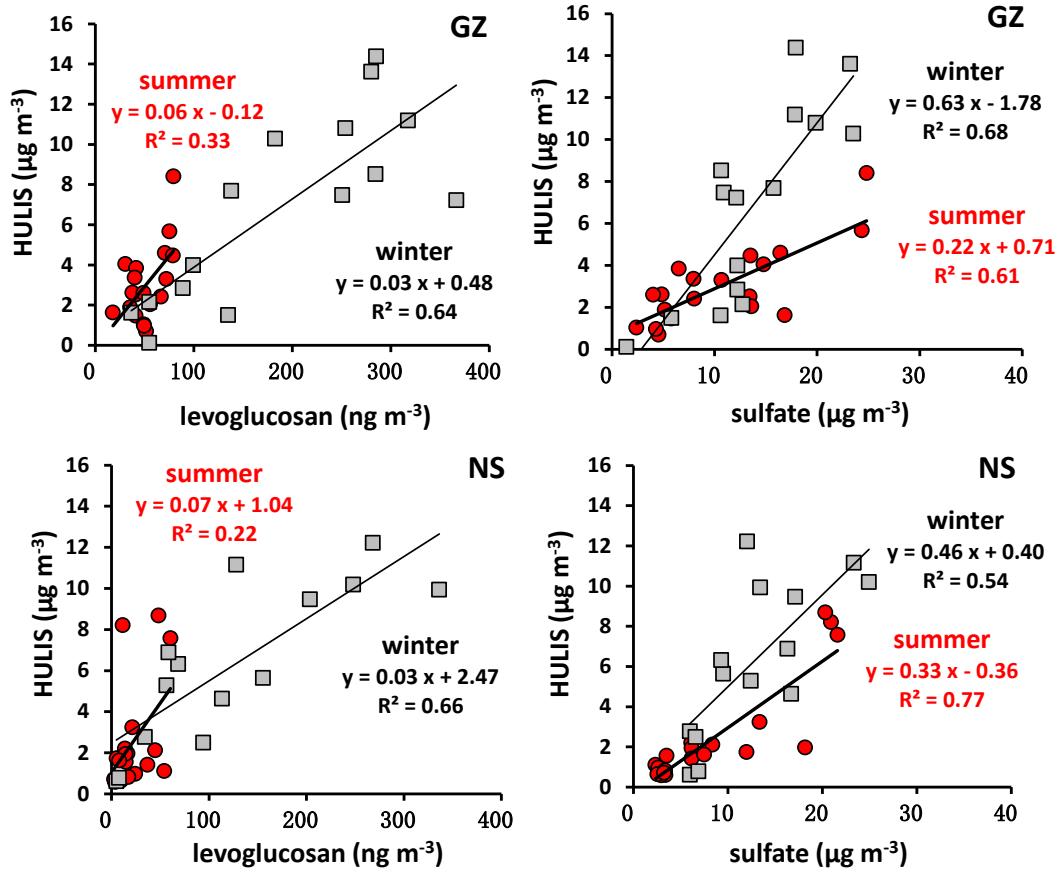
**Fig. S4** Correlation of water-insoluble OC (WISOC) with EC for the GZ and NS samples



**Fig. S5** Average  $\text{PM}_{2.5}$  concentration and composition in GZ and NS in summer and winter. Organic Matter (OM)= $1.4 \times \text{OC}$ . Dust=  $2.2[\text{Al}] + 2.49[\text{Si}] + 1.63[\text{Ca}] + 2.42[\text{Fe}] + 1.94[\text{Ti}]$  (Huang et al., 2014)

#### Reference:

Huang, X. H. H., Bian, Q., Ng, W. M., Louie, P. K. K., and Yu, J. Z.: Characterization of  $\text{PM}_{2.5}$  major components and source investigation in suburban Hong Kong: a one year monitoring study, *Aerosol Air Quality Research*, 14, 237-250, doi:10.4209/aaqr.2013.01.0020, 2014.



**Fig. S6** Correlations of HULIS with levoglucosan and sulfate in summer and winter. The correlations of HULIS vs levoglucosan are much better in winter than in summer, consistent with PMF result that biomass burning source is more important in winter than summer. The correlations of HULIS vs sulfate are similar in winter and in summer, in agreement with the PMF result that secondary process is important in the whole year.

**Table S1.** Concentrations of PM<sub>2.5</sub>, WSOC and HULIS concentrations and influencing air mass origin on individual sampling days

	BT <sup>1</sup>	Guangzhou			Nansha		
		PM <sub>2.5</sub> µg/m <sup>3</sup>	WSOC µgC/m <sup>3</sup>	HULIS µg/m <sup>3</sup>	PM <sub>2.5</sub> µg/m <sup>3</sup>	WSOC µgC/m <sup>3</sup>	HULIS µg/m <sup>3</sup>
2-Jan-09	C	52.3	6.16	7.47	70.0	6.53	12.21
20-Jan-09	T	103.7	10.21	13.61	94.1	8.95	11.16
26-Jan-09		46.6	5.93	4.78	67.5	6.85	9.97
1-Feb-09	T	72.5	6.97	8.52	50.0	4.49	5.64
7-Feb-09	C	126.3	9.92	10.28	81.4	7.40	9.46
13-Feb-09	M				15.9	0.99	0.62
19-Feb-09	M	66.6	3.52	2.15	30.3	2.40	2.77
25-Feb-09	M	33.3	1.73	1.61	24.0	1.39	0.78
3-Mar-09	T	32.3	3.30	2.94	57.4	4.66	4.55
9-Mar-09	C	37.7	2.93	3.15	39.8	3.11	3.76
15-Mar-09	C	70.6	5.99	5.41	53.5	5.91	6.35
27-Mar-09	T	90.8	4.28	4.40	103.0	3.78	4.93
2-Apr-09	T	92.4	5.73	6.55	69.3	4.41	6.14
15-Apr-09	M	79.3	3.41	2.39	42.7	2.72	3.16
26-Apr-09	C	110.3	4.77	3.98	66.0	3.03	3.67
8-May-09	M	40.9	3.37	2.53	35.4	2.81	3.24
20-May-09	M	19.2	1.51	0.70	3.8	1.20	1.11
26-May-09	M	45.9	3.21	2.04	23.6	4.78	2.11
1-Jun-09	T	74.6	8.84	8.40	52.9	6.06	8.20
7-Jun-09	T	48.9	2.90	1.63	35.5	2.22	1.97
13-Jun-09	M	57.8	3.94	3.30	15.2	1.78	1.55
19-Jun-09	M	46.4	4.30	4.05	27.7	1.79	1.74
25-Jun-09	M	29.5	1.95	1.47	11.1	1.20	0.60
1-Jul-09	M	18.7	2.46	1.88	9.6	1.17	0.59
7-Jul-09	M	26.9	3.57	3.85	18.7	2.08	2.21
13-Jul-09	M	52.4	5.60	4.60	18.8	2.04	1.94
19-Jul-09	M	13.6	1.80	1.03	10.7	1.17	0.97
25-Jul-09	M	22.8	2.61	2.62			
31-Jul-09	M	20.8	2.58	2.61	11.1	1.20	0.64
6-Aug-09	M	23.5	1.97	0.97	26.5	1.51	1.42
12-Aug-09	M	35.0	2.35	2.42	13.9	1.26	0.82
14-Aug-09	M				10.3	1.18	0.71
18-Aug-09	M	32.4	3.33	3.37	20.7	1.59	1.62
24-Aug-09	C	47.1	5.60	4.47	64.1	7.12	8.68
30-Aug-09	M	85.3	5.78	5.67	68.6	5.83	7.58
5-Sep-09	M	38.3	5.48	6.96	22.1	2.78	3.35
11-Sep-09	M	37.1	3.27	3.49	36.6	2.44	2.89
17-Sep-09	M	44.2	4.14	3.14	23.9	2.26	2.03
23-Sep-09	C	37.5	3.70	3.45	48.6	4.55	6.04
29-Sep-09	T	46.6	4.64	3.43	39.3	3.78	4.42
5-Oct-09	T	44.6	5.20	4.37	59.8	6.43	8.32
11-Oct-09	T	46.1	6.48	5.05	61.3	7.10	9.27
17-Oct-09	C	77.3	8.40	7.59	69.9	8.14	11.19
23-Oct-09	T	76.8	8.92	8.15	95.3	10.43	14.47
29-Oct-09	T	104.2	9.23	10.95			
4-Nov-09	T	60.7	7.27	7.23	69.2	7.33	9.93
10-Nov-09	M	45.7	4.17	2.84			
16-Nov-09	T	8.5	0.96	0.12			
28-Nov-09	T	131.9	10.71	14.38	47.6	4.62	6.32
4-Dec-09	C	73.8	7.58	10.80	87.9	8.69	10.19
10-Dec-09	C	75.7	5.92	7.69	71.3	4.16	4.63
16-Dec-09	T	21.4	2.38	1.50	26.4	2.38	2.49
22-Dec-09	C	102.8	9.02	11.19	81.9	6.41	6.89
28-Dec-09	C	48.2	3.60	3.99	52.4	3.75	5.28

<sup>1</sup> M=marine; T-transitional; C=continental

**Table S2.** Coefficient of correlation ( $R^2$ ) between HULIS, WSOC\_h and all the species

	GZ, HULIS N=51	NS, HULIS N=49	GZ, WSOC_h N=51	NS, WSOC_h N=49
WSOC_h	0.41	0.39	1.00	1.00
WISOC	<b>0.64</b>	<b>0.59</b>	0.20	0.25
EC	0.39	0.51	0.12	0.31
levoglucosan	<b>0.53</b>	<b>0.53</b>	0.23	0.24
mannosan	<b>0.47</b>	<b>0.42</b>	0.16	0.14
galactosan	<b>0.48</b>	<b>0.42</b>	0.18	0.22
norhopane	0.38	0.19	0.08	0.11
hopane	0.31	0.20	0.05	0.09
NH4+	<b>0.55</b>	<b>0.60</b>	<b>0.44</b>	<b>0.40</b>
NO3-	<b>0.47</b>	<b>0.42</b>	0.26	0.25
SO4=	<b>0.49</b>	<b>0.63</b>	<b>0.56</b>	0.35
C2O4=	<b>0.53</b>	<b>0.33</b>	<b>0.43</b>	<b>0.40</b>
Na+	0.11	0.20	0.07	0.19
Mg2+	0.01	0.07	0.05	0.06
Cl-	0.23	0.12	0.03	0.10
Al	0.02	0.22	0.05	0.10
Si	0.01	0.14	0.03	0.07
K	0.52	0.70	0.40	0.34
Ca	0.02	0.28	0.05	0.16
Ti	0.01	0.19	0.03	0.10
V	0.04	0.01	0.07	0.06
Mn	0.18	0.37	0.12	0.18
Fe	0.04	0.26	0.06	0.11
Ni	0.07	0.11	0.06	0.11
Zn	<b>0.40</b>	<b>0.52</b>	0.23	0.22
Pb	<b>0.43</b>	<b>0.58</b>	0.28	0.25

**Table S3.** Average factor contributions to major PM<sub>2.5</sub> components for the 6-factor solution ( $\mu\text{g m}^{-3}$ )

Factors	GZ site						NS site					
	WSOC_h	EC	OC	sulfate	nitrate	NH <sub>4</sub> <sup>+</sup>	WSOC_h	EC	OC	sulfate	nitrate	NH <sub>4</sub> <sup>+</sup>
F1: Dust	0.08	0.09	0.15	0.78	0	0.10	0.06	0.07	0.11	0.59	0	0.08
F2: Cl <sup>-</sup> and NO <sub>3</sub> <sup>-</sup> dominated factor	0	0.05	2.34	1.48	<b>5.32</b>	<b>2.66</b>	0	0.03	1.57	0.99	<b>3.58</b>	<b>1.79</b>
F3: ship emissions and sea salt aerosols	0.32	<b>0.53</b>	1.29	2.30	0.31	0.79	<b>0.54</b>	<b>0.91</b>	2.23	3.97	0.54	1.36
F4: Secondary process	<b>0.54</b>	0.28	3.17	<b>7.67</b>	0.64	<b>2.90</b>	<b>0.44</b>	0.22	2.57	<b>6.21</b>	0.52	<b>2.35</b>
F5: Biomass burning	<b>0.43</b>	<b>0.64</b>	2.71	0	0.04	0	<b>0.33</b>	<b>0.49</b>	2.06	0	0.03	0
F6: Vehicular emissions	<b>0.82</b>	<b>1.29</b>	2.39	0.36	0.06	0	0.19	<b>0.30</b>	0.56	0.09	0.01	0
Subtotal	2.19	2.88	12.05	12.60	6.37	6.46	1.56	2.03	9.10	11.85	4.68	5.58
Observed conc.	2.31	2.89	12.22	13.39	6.71	6.81	1.46	2.12	9.13	12.15	4.85	5.55