Supplementary Table S1. Summary of the chemical properties of the assigned compositions of HULISs in the size-segregated aerosols.

Sampling Date	Stage No.	Particle Size (μm)	Assigned Peaks	Mean DBE*	Mean AI**	Mean H/C	Mean O/C	Mean N/C	Mean S/C
November 7- December 1	1	0.43-0.65	2888	7.295	0.128	1.261	0.519	0.054	0.03106
	2	0.65-1.1	3035	7.577	0.140	1.236	0.517	0.054	0.03165
	3	1.1-2.1	2873	7.379	0.135	1.248	0.506	0.049	0.03402
	4	2.1-3.3	2063	6.199	0.091	1.332	0.537	0.045	0.04499
	5	3.3-4.7	1801	5.550	0.062	1.393	0.546	0.045	0.05331
	6	4.7-7.0	1694	5.224	0.044	1.435	0.543	0.044	0.05942
	7	7.0-11.0	1468	4.893	0.035	1.480	0.538	0.042	0.06388
	8	>11.0	1516	4.983	0.038	1.476	0.534	0.041	0.06092
January 8-12	1	0.43-0.65	2933	9.156	0.219	1.144	0.456	0.047	0.02387
	2	0.65-1.1	1150	9.418	0.247	1.105	0.454	0.049	0.02257
	3	1.1-2.1	1644	8.925	0.238	1.124	0.449	0.040	0.02446
	4	2.1-3.3	2413	8.049	0.168	1.235	0.453	0.044	0.02979
	5	3.3-4.7	1305	6.079	0.099	1.362	0.504	0.040	0.04868
	6	4.7-7.0	2309	7.009	0.118	1.338	0.447	0.044	0.03731
	7	7.0-11.0	1597	6.143	0.099	1.389	0.465	0.040	0.04578
	8	>11.0	1099	5.642	0.076	1.426	0.491	0.034	0.05087
March 12-16	1	0.43-0.65	1139	4.230	0.067	1.577	0.589	0.052	0.04897
	2	0.65-1.1	1813	6.632	0.092	1.338	0.536	0.050	0.02788
	3	1.1-2.1	1462	6.299	0.079	1.386	0.523	0.044	0.02913
	4	2.1-3.3	1460	5.760	0.057	1.449	0.517	0.034	0.03769
	5	3.3-4.7	1313	5.238	0.035	1.507	0.519	0.035	0.04452
	6	4.7-7.0	1246	4.976	0.030	1.520	0.530	0.035	0.05292
	7	7.0-11.0	1055	4.831	0.030	1.525	0.542	0.034	0.05825
	8	>11.0	1050	4.897	0.038	1.514	0.544	0.030	0.05710

*The double bond equivalent (DBE) value representing the sum of rings and double bonds in each molecule can be calculated from the number of atoms in chemical formulae by the following equation: $DBE=1+C-H/2+N/2$.

**Aromaticity index

Supplementary Table S2. Basic air quality information in the Songdo area provided by the Air Korea website (http://www.airkorea.or.kr) operated by the KME.

	Date	$Temp_{mean}$ (°C)	$Temp_{min}$ (°C)	$Temp_{max}$ (°C)	Instantaneous Wind Speed$_{max}$ (m/s)	Instantaneous wind speed direction$_{max}$ (16 directions)	Wind Speed$_{max}$ (m/s)	Wind speed directionmax (16 directions)	Wind Speed$_{mean}$ (m/s)	Dew-point Temp$_{mean}$ (°C)	Relative Humidity$_{min}$ (%)	Relative Humidity$_{mean}$ (%)	Vapor Pressure$_{mean}$ (hPa)	Atmospheric Pressure$_{mean}$ (hPa)	Ground Temp$_{mean}$ (°C)
Pre-heating period	2017-11-27	1.7	-2.4	7	6	50	3.8	20	2	-5.8	37	58.4	4	1018.1	2.5
	2017-11-28	6	0.8	10	6.2	200	4.3	200	2.1	0.8	56	69.8	6.6	1013.7	4.2
	2017-11-29	2.2	-2.4	8.5	12.7	20	7.4	360	4.9	-7.2	27	52.3	4	1017.8	4.2
	2017-11-30	-2.6	-4.7	0.6	12.7	340	8.3	340	5.8	-16	20	35.8	1.8	1020.3	0
	2017-12-01	-1.6	-6.2	2.6	8.2	20	5.1	270	2.6	-9.7	24	59.1	3.4	1021.9	-0.7
	Average	**1.14**	**-2.98**	**5.74**	**9.16**	**126**	**5.78**	**238**	**3.48**	**-7.58**	**32.8**	**55.08**	**3.96**	**1018.36**	**2.04**
Heating period	2017-01-08	6.3	4.1	8.8	8.4	340	5.9	340	3.7	1.2	57	70.3	6.7	1006	4.4
	2017-01-09	1.9	-2.1	4.4	10.6	320	7.1	340	4.8	-4.1	44	66.3	4.7	1009.1	2.7
	2017-01-10	-2.4	-5.6	0.6	11.6	340	8	340	4.4	-11.6	33	49.6	2.6	1015.4	-1.1
	2017-01-11	-1.9	-6.7	2.3	10.2	360	6.8	340	2.9	-13.2	25	42.6	2.4	1016.3	-1.6
	2017-01-12	-0.7	-4.4	3.1	12.7	290	7.2	340	4.6	-10.2	28	51	3.1	1009.8	-0.8
	Average	**0.64**	**-2.94**	**3.84**	**10.7**	**330**	**7**	**340**	**4.08**	**-7.58**	**37.4**	**55.96**	**3.9**	**1011.32**	**0.72**
Post-heating period	2017-03-12	7.4	5.2	11.4	8.7	340	5.7	340	2.3	2.8	50	73.5	7.6	1012.2	7.7
	2017-03-13	5.7	2.1	10	9.1	360	6.6	340	4	-2.4	32	57.3	5.2	1010.2	6.3
	2017-03-14	5.6	2.2	9.3	16.9	340	8.1	360	5	-6.4	22	42.9	3.8	1010.6	5.2
	2017-03-15	5.3	1.5	10.2	7.6	250	4.9	360	3.1	-6	26	45.1	4	1014.5	4.4
	2017-03-16	5.3	0.9	10.8	7.7	250	5.3	270	2.3	-2.7	27	59.1	5.1	1013.3	4.9
	Average	**5.86**	**2.38**	**10.34**	**10**	**308**	**6.12**	**334**	**3.34**	**-2.94**	**31.4**	**55.58**	**5.14**	**1012.16**	**5.7**

Supplementary Table S3. Chemical properties of the assigned molecular classes of HULISs in the size-segregated aerosols collected during the preheating period.

Stage No. (Particle size, μm)	Class	Mean DBE	Mean AI	Mean H/C	Mean O/C	Mean N/C	Mean S/C
1 (0.43-0.65)	CHO	9.17	0.19	1.13	0.45	0.0000	0.0000
	CHON	8.46	0.20	1.14	0.50	0.0933	0.0000
	CHOS	5.40	0.04	1.41	0.52	0.0000	0.0719
	CHONS	5.23	0.02	1.47	0.62	0.0801	0.0790
2 (0.65-1.1)	CHO	9.75	0.21	1.08	0.46	0.0000	0.0000
	CHON	8.74	0.22	1.12	0.49	0.0919	0.0000
	CHOS	5.52	0.04	1.39	0.53	0.0000	0.0729
	CHONS	5.49	0.02	1.44	0.61	0.0814	0.0790
3 (1.1-2.1)	CHO	9.67	0.22	1.09	0.45	0.0000	0.0000
	CHON	8.52	0.22	1.13	0.47	0.0888	0.0000
	CHOS	5.72	0.05	1.37	0.52	0.0000	0.0726
	CHONS	5.17	0.02	1.46	0.61	0.0838	0.0819
4 (2.1-3.3)	CHO	8.38	0.16	1.15	0.49	0.0000	0.0000
	CHON	7.60	0.19	1.17	0.50	0.0813	0.0000
	CHOS	5.16	0.03	1.42	0.53	0.0000	0.0757
	CHONS	4.54	0.01	1.52	0.62	0.0900	0.0859
5 (3.3-4.7)	CHO	7.42	0.11	1.20	0.53	0.0000	0.0000
	CHON	6.84	0.16	1.22	0.51	0.0851	0.0000
	CHOS	5.10	0.04	1.43	0.52	0.0000	0.0753
	CHONS	4.47	0.01	1.55	0.60	0.0891	0.0850
6 (4.7-7.0)	CHO	7.23	0.09	1.24	0.53	0.0000	0.0000
	CHON	5.92	0.10	1.31	0.53	0.0907	0.0000
	CHOS	5.19	0.04	1.42	0.52	0.0000	0.0748
	CHONS	4.41	0.01	1.56	0.59	0.0878	0.0839
7 (7.0-11.0)	CHO	6.99	0.11	1.24	0.50	0.0000	0.0000
	CHON	5.38	0.09	1.37	0.53	0.0926	0.0000
	CHOS	4.97	0.03	1.46	0.51	0.0000	0.0744
	CHONS	4.30	0.01	1.59	0.58	0.0863	0.0824
8 (>11.0)	CHO	6.60	0.10	1.31	0.49	0.0000	0.0000
	CHON	5.34	0.08	1.40	0.52	0.0884	0.0000
	CHOS	5.12	0.04	1.44	0.51	0.0000	0.0736
	CHONS	4.29	0.01	1.59	0.59	0.0859	0.0827
Overall	CHO	8.15	0.15	1.18	0.49	0.0000	0.0000
	CHON	7.10	0.16	1.23	0.51	0.0890	0.0000
	CHOS	5.27	0.04	1.42	0.52	0.0000	0.0739
	CHONS	4.74	0.01	1.52	0.60	0.0855	0.0825

Supplementary Table S4. Chemical properties of the assigned molecular classes of HULISs in the size-segregated aerosols collected during the main heating period.

Stage No. (Particle size, µm)	Class	Mean DBE	Mean AI	Mean H/C	Mean O/C	Mean N/C	Mean S/C
1 (0.43-0.65)	CHO	11.34	0.31	0.96	0.40	0.0000	0.0000
	CHON	10.70	0.30	1.04	0.40	0.0771	0.0000
	CHOS	5.51	0.05	1.39	0.53	0.0000	0.0728
	CHONS	5.08	0.02	1.48	0.63	0.0804	0.0798
2 (0.65-1.1)	CHO	11.46	0.34	0.90	0.41	0.0000	0.0000
	CHON	10.65	0.32	1.01	0.41	0.0837	0.0000
	CHOS	5.57	0.05	1.38	0.54	0.0000	0.0745
	CHONS	5.07	0.03	1.52	0.64	0.0818	0.0816
3 (1.1-2.1)	CHO	11.42	0.34	0.91	0.40	0.0000	0.0000
	CHON	10.22	0.32	1.02	0.38	0.0753	0.0000
	CHOS	5.21	0.04	1.40	0.53	0.0000	0.0770
	CHONS	3.98	0.01	1.59	0.70	0.0902	0.0902
4 (2.1-3.3)	CHO	10.18	0.26	1.05	0.41	0.0000	0.0000
	CHON	9.57	0.25	1.15	0.38	0.0754	0.0000
	CHOS	5.41	0.04	1.39	0.53	0.0000	0.0763
	CHONS	4.84	0.01	1.51	0.59	0.0827	0.0819
5 (3.3-4.7)	CHO	8.74	0.23	1.09	0.44	0.0000	0.0000
	CHON	7.72	0.20	1.20	0.43	0.0833	0.0000
	CHOS	4.87	0.03	1.46	0.51	0.0000	0.0767
	CHONS	4.45	0.01	1.57	0.59	0.0878	0.0866
6 (4.7-7.0)	CHO	8.94	0.21	1.14	0.41	0.0000	0.0000
	CHON	8.59	0.20	1.26	0.35	0.0722	0.0000
	CHOS	5.41	0.04	1.41	0.51	0.0000	0.0736
	CHONS	5.16	0.02	1.50	0.54	0.0784	0.0768
7 (7.0-11.0)	CHO	8.28	0.23	1.15	0.42	0.0000	0.0000
	CHON	7.87	0.19	1.28	0.35	0.0743	0.0000
	CHOS	5.07	0.04	1.44	0.51	0.0000	0.0755
	CHONS	4.49	0.01	1.57	0.55	0.0829	0.0811
8 (>11.0)	CHO	8.22	0.20	1.18	0.42	0.0000	0.0000
	CHON	7.03	0.15	1.31	0.40	0.0766	0.0000
	CHOS	4.85	0.03	1.47	0.50	0.0000	0.0745
	CHONS	4.04	0.01	1.62	0.59	0.0866	0.0852
Overall	CHO	9.82	0.27	1.05	0.42	0.0000	0.0000
	CHON	9.04	0.24	1.16	0.39	0.0772	0.0000
	CHOS	5.24	0.04	1.42	0.52	0.0000	0.0751
	CHONS	4.64	0.02	1.55	0.60	0.0838	0.0829

Supplementary Table S5. Chemical properties of the assigned molecular classes of HULISs in the size-segregated aerosols collected during the postheating period.

Stage No. (Particle size, μm)	Class	Mean DBE	Mean AI	Mean H/C	Mean O/C	Mean N/C	Mean S/C
1 (0.43-0.65)	CHO	7.00	0.15	1.21	0.46	0.0000	0.0000
	CHON	5.43	0.15	1.33	0.55	0.1080	0.0000
	CHOS	2.77	0.01	1.78	0.56	0.0000	0.0773
	CHONS	3.36	0.01	1.75	0.76	0.0843	0.0843
2 (0.65-1.1)	CHO	8.12	0.17	1.12	0.50	0.0000	0.0000
	CHON	7.97	0.13	1.23	0.49	0.0830	0.0000
	CHOS	4.22	0.00	1.54	0.57	0.0000	0.0751
	CHONS	4.03	0.00	1.64	0.67	0.0806	0.0800
3 (1.1-2.1)	CHO	8.03	0.16	1.17	0.48	0.0000	0.0000
	CHON	7.58	0.11	1.28	0.48	0.0766	0.0000
	CHOS	4.15	0.01	1.56	0.56	0.0000	0.0745
	CHONS	3.66	0.00	1.70	0.65	0.0810	0.0803
4 (2.1-3.3)	CHO	7.87	0.16	1.20	0.47	0.0000	0.0000
	CHON	6.85	0.07	1.38	0.47	0.0671	0.0000
	CHOS	4.54	0.01	1.53	0.53	0.0000	0.0713
	CHONS	3.76	0.00	1.69	0.62	0.0823	0.0792
5 (3.3-4.7)	CHO	6.99	0.11	1.30	0.48	0.0000	0.0000
	CHON	6.18	0.04	1.45	0.47	0.0683	0.0000
	CHOS	4.66	0.01	1.52	0.53	0.0000	0.0715
	CHONS	3.98	0.00	1.68	0.58	0.0800	0.0766
6 (4.7-7.0)	CHO	7.36	0.12	1.25	0.49	0.0000	0.0000
	CHON	5.66	0.05	1.48	0.50	0.0750	0.0000
	CHOS	4.59	0.01	1.53	0.52	0.0000	0.0714
	CHONS	4.03	0.01	1.66	0.57	0.0818	0.0778
7 (7.0-11.0)	CHO	7.77	0.14	1.18	0.49	0.0000	0.0000
	CHON	5.17	0.04	1.49	0.52	0.0834	0.0000
	CHOS	4.70	0.01	1.52	0.52	0.0000	0.0715
	CHONS	3.73	0.00	1.69	0.60	0.0854	0.0810
8 (>11.0)	CHO	7.73	0.14	1.21	0.49	0.0000	0.0000
	CHON	5.11	0.06	1.47	0.51	0.0858	0.0000
	CHOS	4.64	0.02	1.52	0.53	0.0000	0.0722
	CHONS	3.56	0.00	1.71	0.62	0.0871	0.0832
Overall	CHO	7.61	0.14	1.20	0.48	0.0000	0.0000
	CHON	6.25	0.08	1.39	0.50	0.0809	0.0000
	CHOS	4.28	0.01	1.56	0.54	0.0000	0.0731
	CHONS	3.77	0.00	1.69	0.63	0.0828	0.0803

Supplementary Table S6. Chemical properties of the assigned compound classes of HULISs in the size-segregated aerosols collected during the preheating period.

Stage No. (Particle size, μm)	Class	Mean DBE	Mean AI	Mean H/C	Mean O/C	Mean N/C	Mean S/C
1 (0.43-0.65)	Lignins	9.08	0.16	1.10	0.45	0.0534	0.0217
	Tannins	5.83	0.01	1.13	0.82	0.0692	0.0486
	Unsaturated hydrocarbons	21.00	0.71	0.77	0.00	0.0866	0.0330
	Proteins	3.69	0.00	1.72	0.51	0.0716	0.0392
	Carbohydrates	2.67	0.00	1.75	0.89	0.0895	0.0671
	Condensed aromatics	11.79	0.76	0.58	0.35	0.0668	0.0014
	Lipids	3.82	0.00	1.72	0.27	0.0077	0.0309
2 (0.65-1.1)	Lignins	9.26	0.17	1.10	0.45	0.0531	0.0221
	Tannins	5.89	0.01	1.11	0.82	0.0659	0.0512
	Unsaturated hydrocarbons	20.50	0.78	0.73	0.00	0.1455	0.0364
	Proteins	3.68	0.00	1.72	0.51	0.0732	0.0407
	Carbohydrates	2.68	0.00	1.74	0.90	0.0862	0.0695
	Condensed aromatics	12.89	0.77	0.58	0.32	0.0806	0.0053
	Lipids	3.70	0.00	1.72	0.27	0.0112	0.0395
3 (1.1-2.1)	Lignins	9.09	0.17	1.10	0.44	0.0482	0.0242
	Tannins	5.76	0.01	1.12	0.82	0.0623	0.0560
	Unsaturated hydrocarbons	0.00	0.00	0.00	0.00	0.0000	0.0000
	Proteins	3.69	0.00	1.71	0.50	0.0792	0.0457
	Carbohydrates	2.55	0.00	1.75	0.89	0.0742	0.0778
	Condensed aromatics	11.57	0.74	0.57	0.36	0.0608	0.0000
	Lipids	3.84	0.01	1.70	0.26	0.0247	0.0293
4 (2.1-3.3)	Lignins	8.10	0.13	1.14	0.46	0.0397	0.0320
	Tannins	5.52	0.01	1.14	0.82	0.0620	0.0677
	Unsaturated hydrocarbons	21.00	0.77	0.71	0.07	0.1429	0.0000
	Proteins	3.62	0.00	1.72	0.50	0.0779	0.0529
	Carbohydrates	2.41	0.00	1.77	0.89	0.0661	0.0849
	Condensed aromatics	10.85	0.75	0.57	0.38	0.0631	0.0103
	Lipids	3.65	0.01	1.71	0.26	0.0165	0.0402
5 (3.3-4.7)	Lignins	7.45	0.10	1.18	0.46	0.0380	0.0396
	Tannins	5.42	0.01	1.15	0.83	0.0629	0.0768
	Unsaturated hydrocarbons	21.00	0.77	0.71	0.07	0.1429	0.0000
	Proteins	3.60	0.00	1.72	0.49	0.0785	0.0577
	Carbohydrates	2.38	0.00	1.78	0.90	0.0692	0.0881
	Condensed aromatics	10.67	0.76	0.56	0.37	0.0604	0.0222
	Lipids	3.51	0.01	1.72	0.27	0.0214	0.0479
6 (4.7-7.0)	Lignins	7.16	0.08	1.21	0.46	0.0343	0.0479
	Tannins	5.23	0.01	1.15	0.85	0.0666	0.0819
	Unsaturated hydrocarbons	27.00	0.57	0.84	0.07	0.0000	0.0000
	Proteins	3.51	0.00	1.73	0.49	0.0785	0.0593
	Carbohydrates	2.39	0.00	1.78	0.90	0.0703	0.0913
	Condensed aromatics	11.25	0.79	0.50	0.37	0.0503	0.0490
	Lipids	3.65	0.00	1.71	0.27	0.0257	0.0508
7 (7.0-11.0)	Lignins	6.87	0.07	1.23	0.45	0.0321	0.0534
	Tannins	5.11	0.01	1.18	0.84	0.0634	0.0896
	Unsaturated hydrocarbons	0.00	0.00	0.00	0.00	0.0000	0.0000
	Proteins	3.51	0.00	1.73	0.49	0.0777	0.0604
	Carbohydrates	2.34	0.00	1.79	0.91	0.0692	0.0957
	Condensed aromatics	11.33	0.84	0.51	0.28	0.0667	0.0889
	Lipids	3.66	0.00	1.71	0.27	0.0290	0.0532
8 (>11.0)	Lignins	6.94	0.07	1.24	0.45	0.0315	0.0507
	Tannins	5.09	0.00	1.19	0.83	0.0600	0.0883
	Unsaturated hydrocarbons	0.00	0.00	0.00	0.00	0.0000	0.0000
	Proteins	3.60	0.00	1.72	0.49	0.0745	0.0566
	Carbohydrates	2.36	0.00	1.78	0.90	0.0721	0.0937
	Condensed aromatics	11.75	0.82	0.48	0.29	0.0500	0.1000
	Lipids	3.70	0.01	1.70	0.27	0.0237	0.0482

Supplementary Table S7. Chemical properties of the assigned compound classes of HULISs in the size-segregated aerosols collected during the main heating period.

Stage No. (Particle size, μm)	Class	Mean DBE	Mean AI	Mean H/C	Mean O/C	Mean N/C	Mean S/C
1 (0.43-0.65)	Lignins	10.39	0.23	1.06	0.41	0.0472	0.0148
	Tannins	5.64	0.01	1.16	0.78	0.0582	0.0568
	Unsaturated hydrocarbons	8.00	0.33	1.30	0.10	0.0000	0.0000
	Proteins	3.65	0.00	1.72	0.53	0.0708	0.0463
	Carbohydrates	2.53	0.00	1.77	0.86	0.0689	0.0871
	Condensed aromatics	13.11	0.74	0.57	0.31	0.0517	0.0008
	Lipids	4.25	0.00	1.69	0.28	0.0161	0.0289
2 (0.65-1.1)	Lignins	10.28	0.25	1.04	0.41	0.0488	0.0136
	Tannins	5.83	0.01	1.12	0.77	0.0640	0.0429
	Unsaturated hydrocarbons	14.60	0.62	0.93	0.05	0.1183	0.0572
	Proteins	3.64	0.00	1.72	0.53	0.0726	0.0435
	Carbohydrates	2.51	0.00	1.79	0.86	0.0657	0.0908
	Condensed aromatics	15.18	0.75	0.58	0.28	0.0616	0.0037
	Lipids	5.50	0.02	1.56	0.27	0.0670	0.0132
3 (1.1-2.1)	Lignins	9.96	0.25	1.04	0.40	0.0412	0.0142
	Tannins	5.23	0.01	1.21	0.76	0.0473	0.0630
	Unsaturated hydrocarbons	17.00	0.45	1.09	0.10	0.0233	0.0000
	Proteins	3.44	0.00	1.72	0.56	0.0777	0.0499
	Carbohydrates	2.40	0.00	1.79	0.86	0.0643	0.0988
	Condensed aromatics	13.24	0.72	0.57	0.28	0.0333	0.0000
	Lipids	3.00	0.02	1.78	0.27	0.0313	0.0294
4 (2.1-3.3)	Lignins	9.50	0.20	1.12	0.40	0.0438	0.0191
	Tannins	5.20	0.01	1.20	0.80	0.0540	0.0729
	Unsaturated hydrocarbons	18.00	0.70	0.90	0.02	0.1508	0.0613
	Proteins	3.80	0.00	1.70	0.49	0.0763	0.0496
	Carbohydrates	2.40	0.00	1.78	0.87	0.0618	0.0928
	Condensed aromatics	12.63	0.73	0.57	0.31	0.0365	0.0000
	Lipids	4.94	0.02	1.65	0.26	0.0406	0.0205
5 (3.3-4.7)	Lignins	7.85	0.14	1.16	0.43	0.0351	0.0334
	Tannins	4.88	0.01	1.21	0.81	0.0576	0.0837
	Unsaturated hydrocarbons	17.20	0.44	1.11	0.05	0.0260	0.0313
	Proteins	3.63	0.00	1.71	0.49	0.0768	0.0588
	Carbohydrates	2.26	0.00	1.80	0.89	0.0668	0.0995
	Condensed aromatics	11.00	0.76	0.58	0.34	0.0526	0.0000
	Lipids	4.10	0.01	1.71	0.25	0.0283	0.0367
6 (4.7-7.0)	Lignins	8.62	0.16	1.18	0.39	0.0419	0.0267
	Tannins	5.09	0.01	1.21	0.80	0.0581	0.0855
	Unsaturated hydrocarbons	27.00	0.57	0.84	0.07	0.0000	0.0000
	Proteins	3.71	0.00	1.71	0.47	0.0751	0.0527
	Carbohydrates	2.32	0.00	1.79	0.88	0.0662	0.0947
	Condensed aromatics	12.48	0.73	0.57	0.30	0.0317	0.0000
	Lipids	5.22	0.02	1.64	0.25	0.0486	0.0194
7 (7.0-11.0)	Lignins	7.92	0.15	1.20	0.39	0.0361	0.0326
	Tannins	4.79	0.01	1.21	0.82	0.0594	0.0911
	Unsaturated hydrocarbons	27.00	0.57	0.84	0.07	0.0000	0.0000
	Proteins	3.67	0.00	1.72	0.46	0.0744	0.0581
	Carbohydrates	2.30	0.00	1.80	0.89	0.0664	0.1035
	Condensed aromatics	12.00	0.76	0.55	0.29	0.0287	0.0045
	Lipids	4.82	0.02	1.66	0.25	0.0432	0.0282
8 (>11.0)	Lignins	7.52	0.12	1.20	0.41	0.0259	0.0373
	Tannins	4.84	0.01	1.21	0.80	0.0666	0.0860
	Unsaturated hydrocarbons	27.00	0.57	0.84	0.07	0.0000	0.0000
	Proteins	3.64	0.00	1.71	0.48	0.0747	0.0589
	Carbohydrates	2.27	0.00	1.80	0.88	0.0632	0.1030
	Condensed aromatics	13.29	0.73	0.57	0.22	0.0179	0.0000
	Lipids	4.43	0.01	1.69	0.27	0.0169	0.0346

Supplementary Table S8. Chemical properties of the assigned compound classes of HULISs in the size-segregated aerosols collected during the postheating period.

Stage No. (Particle size, μm)	Class	Mean DBE	Mean AI	Mean H/C	Mean O/C	Mean N/C	Mean S/C
1 (0.43-0.65)	Lignins	7.19	0.16	1.20	0.46	0.0521	0.0094
	Tannins	4.36	0.05	1.30	0.79	0.0910	0.0348
	Unsaturated hydrocarbons	16.00	0.41	1.21	0.04	0.0625	0.0288
	Proteins	3.15	0.00	1.79	0.59	0.0770	0.0455
	Carbohydrates	2.58	0.00	1.75	0.84	0.0738	0.0840
	Condensed aromatics	0.00	0.00	0.00	0.00	0.0000	0.0000
	Lipids	4.67	0.01	1.70	0.16	0.0185	0.0370
2 (0.65-1.1)	Lignins	8.52	0.13	1.15	0.47	0.0532	0.0140
	Tannins	5.48	0.01	1.21	0.78	0.0495	0.0410
	Unsaturated hydrocarbons	35.00	0.60	0.83	0.00	0.0000	0.0172
	Proteins	3.84	0.00	1.71	0.52	0.0708	0.0382
	Carbohydrates	2.61	0.00	1.76	0.88	0.0777	0.0770
	Condensed aromatics	13.13	0.74	0.56	0.35	0.0453	0.0000
	Lipids	4.04	0.01	1.71	0.26	0.0192	0.0161
3 (1.1-2.1)	Lignins	8.43	0.12	1.16	0.47	0.0440	0.0142
	Tannins	5.27	0.01	1.24	0.77	0.0444	0.0435
	Unsaturated hydrocarbons	27.00	0.57	0.84	0.07	0.0000	0.0000
	Proteins	3.74	0.00	1.72	0.52	0.0711	0.0409
	Carbohydrates	2.48	0.00	1.78	0.87	0.0739	0.0797
	Condensed aromatics	12.33	0.76	0.56	0.31	0.0370	0.0000
	Lipids	4.14	0.01	1.71	0.26	0.0316	0.0161
4 (2.1-3.3)	Lignins	8.00	0.09	1.20	0.46	0.0276	0.0237
	Tannins	5.00	0.01	1.25	0.78	0.0521	0.0633
	Unsaturated hydrocarbons	24.00	0.56	0.87	0.05	0.0000	0.0111
	Proteins	3.69	0.00	1.72	0.50	0.0702	0.0462
	Carbohydrates	2.40	0.00	1.79	0.88	0.0656	0.0844
	Condensed aromatics	13.38	0.73	0.52	0.35	0.0139	0.0000
	Lipids	4.00	0.00	1.73	0.26	0.0215	0.0218
5 (3.3-4.7)	Lignins	7.45	0.06	1.24	0.46	0.0244	0.0331
	Tannins	4.89	0.01	1.25	0.80	0.0570	0.0721
	Unsaturated hydrocarbons	25.00	0.58	0.87	0.05	0.0263	0.0132
	Proteins	3.65	0.00	1.73	0.50	0.0718	0.0502
	Carbohydrates	2.36	0.00	1.79	0.88	0.0635	0.0873
	Condensed aromatics	10.33	0.78	0.48	0.44	0.0370	0.0000
	Lipids	4.47	0.01	1.69	0.26	0.0237	0.0235
6 (4.7-7.0)	Lignins	7.18	0.06	1.24	0.46	0.0230	0.0414
	Tannins	5.00	0.01	1.23	0.81	0.0539	0.0792
	Unsaturated hydrocarbons	19.67	0.48	1.05	0.08	0.0667	0.0333
	Proteins	3.63	0.00	1.73	0.49	0.0730	0.0550
	Carbohydrates	2.32	0.00	1.80	0.88	0.0644	0.0912
	Condensed aromatics	11.00	0.75	0.46	0.38	0.0000	0.0000
	Lipids	3.93	0.01	1.72	0.26	0.0237	0.0349
7 (7.0-11.0)	Lignins	7.09	0.06	1.24	0.46	0.0158	0.0466
	Tannins	4.94	0.01	1.22	0.80	0.0621	0.0793
	Unsaturated hydrocarbons	27.00	0.55	0.87	0.08	0.0000	0.0000
	Proteins	3.58	0.00	1.73	0.49	0.0735	0.0572
	Carbohydrates	2.26	0.00	1.81	0.89	0.0661	0.0955
	Condensed aromatics	11.00	0.75	0.46	0.38	0.0000	0.0000
	Lipids	3.88	0.01	1.72	0.27	0.0196	0.0400
8 (>11.0)	Lignins	7.34	0.07	1.21	0.47	0.0127	0.0434
	Tannins	4.97	0.01	1.22	0.80	0.0549	0.0844
	Unsaturated hydrocarbons	27.00	0.57	0.84	0.07	0.0000	0.0000
	Proteins	3.58	0.00	1.73	0.49	0.0725	0.0582
	Carbohydrates	2.27	0.00	1.80	0.89	0.0614	0.0960
	Condensed aromatics	9.50	0.81	0.51	0.41	0.0556	0.0000
	Lipids	3.65	0.01	1.74	0.26	0.0178	0.0367

Supplementary Table S9. PM10 and PM2.5 levels measured in the Songdo area, South Korea, provided by Air Korea.

PM$_{10}$ ($\mu g/m^3$)

Pre-heating period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	1	Ave*
September 2017	34	36	35	64	33	34	39	56	45	47	53	31	40	39	23	25	43	32	21	30	43	47	20	18	31	40	43	63	46	18	21	38.2

Heating period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	Ave*
January 2018	34	36	25	38	44	38	51	50	28	24	23	18	42	56	68	113	82	72	45	93	46	38	17	21	17	25	38	24	37	34	36	28.6

Post-heating period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	Ave*
March 2018	37	54	41	24	8	29	29	33	47	32	56	88	74	21	21	15	27	49	28	19	15	29	46	75	94	65	73	45	41	51	45	43.8

PM$_{2.5}$ ($\mu g/m^3$)

Pre-heating period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	1	Ave*
September 2017	18	21	22	53	21	21	24	25	23	24	10	17	25	13	10	13	27	17	9	18	27	27	9	10	16	24	28	45	19	5	11	21.6

Heating period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	Ave*
January 2018	18	22	13	23	30	27	40	37	13	10	14	11	34	45	50	96	71	52	23	68	30	25	6	6	7	14	25	13	12	17	18	17.0

Post-heating period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	Ave*
March 2018	26	18	22	16	5	17	17	25	36	23	43	67	58	12	14	9	17	40	21	8	8	18	36	61	82	47	54	33	31	32	27	32.0

*The average values were calculated with the size-segregated sampling periods.