Supplementary materials for: Biomass burning contribution to Beijing aerosol

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Section 1. Summary of the published levoglucosan to K^+ ratios (L/K) and the levoglucosan to mannosan ratios (L/M).

Biomass type	Combustion style	levoglucosan	K^+	mannosan	L/K	L/M	Reference
US hard wood							
Red maple	fireplace	9.28	1.24	0.28	7.51	33.02	Fine et al., 2001[a]
Northern red oak	fireplace	14.72	1.00	0.42	14.71	35.46	Fine et al., 2001[a]
Paper birch	fireplace	9.51	0.98	0.11	9.74	83.43	Fine et al., 2001[a]
Yellow poplar	fireplace	13.26	0.73	1.24	18.27	10.67	Fine et al., 2002[a]
White ash	fireplace	7.60	1.75	0.59	4.34	12.88	Fine et al., 2002[a]
Sweetgum	fireplace	10.09	0.80	0.55	12.65	18.38	Fine et al., 2002[a]
Mockernut hickory	fireplace	11.81	0.20	0.48	59.06	24.73	Fine et al., 2002[a]
White oak	fireplace	7.41	0.90	0.57	8.20	12.89	Fine et al., 2004a[a]
Sugar maple	fireplace	17.37	0.66	0.88	26.44	19.76	Fine et al., 2004a[a]
Black oak	fireplace	17.78	1.03	0.76	17.30	23.40	Fine et al., 2004a[a]
American beech	fireplace	5.62	0.40	0.33	14.04	16.89	Fine et al., 2004a[a]
Black cherry	fireplace	22.65	1.90	1.15	11.91	19.65	Fine et al., 2004a[a]
Quaking aspen	fireplace	12.54	0.45	0.87	28.12	14.46	Fine et al., 2004a[a]
Red maple	stove	12.66	1.68	0.66	7.54	19.27	Fine et al., 2004b[a]
White oak	stove	6.93	1.51	0.31	4.58	22.70	Fine et al., 2004b[a]
White oak (catalyst)	stove	5.93	2.74	0.23	2.16	26.05	Fine et al., 2004b[a]
Sugar maple	stove	10.76	1.01	0.66	10.61	16.31	Fine et al., 2004b[a]
Oak	open chamber	500.00		18.00		27.78	Engling et al., 2006[b]
Oak	open chamber	252.00		7.80		32.31	Engling et al., 2006[b]
Cottonwood	open chamber	276.00		20.00		13.80	Engling et al., 2006[b]
Oak	fireplace	0.71	0.03		21.40		Schauer et al., 2001[c]
Eucalyptus	fireplace	1.94	0.07		28.21		Schauer et al., 2001[c]

US softwood							
Eastern white pine	fireplace	3.84	0.44	0.66	8.75	5.81	Fine et al., 2001[a]
Eastern hemlock	fireplace	9.76	1.32	2.62	7.38	3.73	Fine et al., 2001[a]
Balsam fir	fireplace	8.66	1.48	1.85	5.85	4.68	Fine et al., 2001[a]
Loblolly pine	fireplace	3.65	0.44	0.81	8.40	4.53	Fine et al., 2002[a]
Slash pine	fireplace	4.72	0.65	0.96	7.23	4.91	Fine et al., 2002[a]
White spruce	fireplace	12.72	0.42	3.23	30.22	3.94	Fine et al., 2004a[a]
Douglar fir	fireplace	26.15	0.37	5.89	71.45	4.44	Fine et al., 2004a[a]
Ponderosa pine	fireplace	6.40	0.44	1.89	14.61	3.38	Fine et al., 2004a[a]
Pinyon pine	fireplace	0.80	0.16	0.12	5.01	6.67	Fine et al., 2004a[a]
Pine	fireplace	1.38	0.03		52.25		Schauer et al., 2001[c]
Loblolly pine	stove	11.04	0.46	2.02	24.09	5.46	Fine et al., 2004b[a]
Douglas fir	stove	31.80	1.17	9.15	27.16	3.47	Fine et al., 2004b[a]
Douglas fir (catalyst)	stove	25.67	0.62	4.41	41.34	5.82	Fine et al., 2004b[a]
Ponderosa pine	open chamber	80.00		26.00		3.08	Engling et al., 2006[b]
Ponderosa pine	open chamber	36.00		7.20		5.00	Engling et al., 2006[b]
Ponderosa pine	open chamber	248.00		97.00		2.56	Engling et al., 2006[b]
Ponderosa pine	open chamber	539.00		175.00		3.08	Engling et al., 2006[b]
Ponderosa pine	open chamber	3.55	0.16		22.17		Hays et al., 2002[a]
Western hemlock	open chamber	3.18	0.69		4.61		Hays et al., 2002[a]
Europen hardwood							
Beech	stove	4.10	0.21	0.28	19.25	14.64	Schmidl et al., 2008a[a]
Oak	stove	13.30	0.41	0.92	32.13	14.46	Schmidl et al., 2008a[a]
Eucalyptus	chamber	14.70		0.43		34.19	Gonçalves et al., 2010[a]
Cork oak	chamber	6.80		0.27		25.19	Gonçalves et al., 2010[a]

Golden wattle	chamber	1.90		0.17		11.18	Gonçalves et al., 2010[a]
Beech	house	2286.78		155.02		14.75	Bari et al., 2009[a]
Europen softwood							
Spruce	stove	10.70	0.16	3.00	66.88	3.57	Schmidl et al., 2008a[a]
Larch	stove	15.10	0.07	3.90	206.85	3.87	Schmidl et al., 2008a[a]
Briquettes	stove	10.10	0.19	4.00	53.44	2.53	Schmidl et al., 2008a[a]
Maritime pine	chamber	3.80		1.30		2.92	Gonçalves et al., 2010[a]
Pine	chamber	1.20	0.0046	0.32	260.87	3.75	Iinuma et al., 2007[c]
Pine with green needles	chamber	0.85	0.084	0.13	10.12	6.54	Iinuma et al., 2007[c]
Spruce with green needles	chamber	0.99	0.028	0.21	35.36	4.71	Iinuma et al., 2007[c]
US Needle							
Needle	open chamber	5.64	1.05	1.35	5.37	4.18	Sullivan et al., 2008[d]
Needle	open chamber	25.53	4.66	8.32	5.48	3.07	Sullivan et al., 2008[d]
Needle	open chamber	28.32	4.37	6.72	6.48	4.21	Sullivan et al., 2008[d]
Needle	open chamber	4.76	0.70	1.64	6.80	2.90	Sullivan et al., 2008[d]
Needle	open chamber	30.84	2.95	8.94	10.45	3.45	Sullivan et al., 2008[d]
Needle	open chamber	12.70	5.53	8.97	2.30	1.42	Sullivan et al., 2008[d]
Needle	open chamber	25.01	7.19	14.25	3.48	1.76	Sullivan et al., 2008[d]
Needle	open chamber	8.73	1.66	2.10	5.26	4.16	Sullivan et al., 2008[d]
Needle	open chamber	10.53	1.07	6.42	9.84	1.64	Sullivan et al., 2008[d]
Needle	open chamber	7.31	3.14	1.35	2.33	5.41	Sullivan et al., 2008[d]
Needle	open chamber	186.00		50.00		3.72	Engling et al., 2006[b]
Needle	open chamber	98.00		15.00		6.53	Engling et al., 2006[b]
Needle	open chamber	530.00		144.00		3.68	Engling et al., 2006[b]

Needle	open chamber	247.00		66.00		3.74	Engling et al., 2006[b]
US grass							
Grass	open chamber	6.66	12.21	0.51	0.55	13.06	Sullivan et al., 2008[d]
Grass	open chamber	8.73	29.34	0.62	0.30	14.08	Sullivan et al., 2008[d]
Grass	open chamber	4.77	58.51	0.52	0.08	9.17	Sullivan et al., 2008[d]
Grass	open chamber	16.85	4.44	0.92	3.80	18.32	Sullivan et al., 2008[d]
Grass	open chamber	7.98	0.84	0.36	9.50	22.17	Sullivan et al., 2008[d]
Grass	open chamber	20.36	2.25	0.52	9.05	39.15	Sullivan et al., 2008[d]
Grass	open chamber	18.88	301.98	1.64	0.06	11.51	Sullivan et al., 2008[d]
US duff							
Duff	open chamber	12.59	2.20	11.60	5.72	1.09	Sullivan et al., 2008[d]
Duff	open chamber	14.67		6.32		2.32	Sullivan et al., 2008[d]
Duff	open chamber	23.71	1.09	17.79	21.75	1.33	Sullivan et al., 2008[d]
Duff	open chamber	14.46	1.12	14.64	12.91	0.99	Sullivan et al., 2008[d]
Duff	open chamber	8.41	0.37	4.37	22.73	1.92	Sullivan et al., 2008[d]
Duff	open chamber	11.94	0.70	11.97	17.06	1.00	Sullivan et al., 2008[d]
Duff	open chamber	9.87		5.86		1.68	Sullivan et al., 2008[d]
Duff	open chamber	525.00		216.00		2.43	Engling et al., 2006[c]
Asia Rice straw							
Rice straw (Taiwan)	open chamber	1.97	6.84	0.16	0.29	12.31	Sullivan et al., 2008[d]
Rice straw (Taiwan)	open chamber	28.76	39.49	1.53	0.73	18.80	Sullivan et al., 2008[d]
Rice straw (Taiwan)	open chamber	33.02	36.29	0.60	0.91	55.03	Sullivan et al., 2008[d]
Rice straw (Taiwan)	open chamber	12.02	25.26	0.85	0.48	14.14	Sullivan et al., 2008[d]

Rice straw (Taiwan)	open chamber	25.69	21.09	0.76	1.22	33.80	Sullivan et al., 2008[d]
Rice straw (Taiwan)	open chamber	25.05	38.58	0.81	0.65	30.93	Sullivan et al., 2008[d]
Rice straw (Taiwan)	open chamber	7.44	25.66	0.15	0.29	49.60	Sullivan et al., 2008[d]
Rice straw (Bangladesh)	stove	1.83	2.50	0.04	0.73	41.59	Sheesley et al., 2003[a]
Rice straw (Thailand)	open burning	56.20	50.00		1.12		Oanh et al., 2011[a]
Rice straw (Taiwan)	filed burning	0.932	3.908		0.24		Engling et al., 2009[d]
Rice straw (Taiwan)	filed burning	1.67	3.167		0.53		Engling et al., 2009[d]
Rice straw (Taiwan)	filed burning	2.527	4.452		0.57		Engling et al., 2009[d]
Rice straw (Taiwan)	filed burning	1.200	4.172		0.29		Engling et al., 2009[d]
Rice straw (Taiwan)	filed burning	3.1364		0.123		25.48	Engling et al., 2009[d]

a wt% of PM mass

b μg/mg-OC

c g/kg-fuel

d $\mu g/m^3$

Section 2. Estimation of the uncertainties of OC, EC, WSOC, and water soluble ions.

Parallel analysis of OC, EC and water-soluble ions was not performed in the present study. However, their uncertainties could be estimated based on our previous work about the measurement method of ambient aerosol. Results from the inter-comparison of thermal-optical methods suggested that uncertainty of the total carbon (TC) quantified by the DRI carbon analyzer was about 3% (Cheng et al., 2011, 2012a). On the other hand, the EC uncertainty could be calculated using the EC values measured by the bare and denuded quartz filters (the use of charcoal denuder will not influence the EC results, since EC exists only in the particle phase). Using this approach, the EC uncertainty was estimated to be about 4% based on the results from Cheng et al. (2010). Comparison of the TC uncertainty and the EC uncertainty indicates that the OC uncertainty should be comparable with EC. Therefore, a value of 5% was used as the uncertainty of OC and EC in the PMF model. Similarly, the uncertainty of SO_4^{2-} and K^+ could also be calculated using their values measured by the bare and denuded quartz filters (the use of charcoal denuder will not influence the SO_4^{2-} and K^+ results, since they exists only in the particle phase). Based on the results from Cheng et al. (2012b), the uncertainty of SO_4^{2-} and K^+ was estimated to be about 3% and 4%, respectively. In the PMF model, a value of 5% was used as the uncertainty of SO_4^{2-} and K^+. Oxalate was not measured by Cheng et al. (2012b), thus, it is assumed that the oxalate uncertainty was twice the value of that for K^+ (i.e., 10%).

When performing the WSOC analysis at Georgia Institute of Technology, four ambient samples and eight sucrose solutions were analyzed twice. The results showed that the WSOC uncertainty was about 2% and 3% for the ambient samples and the sucrose solutions, respectively. In the PMF analysis, a value of 5% was used as the WSOC uncertainty.

References

Cheng Y., He K. B., Duan F. K., Zheng M., Ma Y. L., Tan J. H., and Du Z. Y.: Improved Measurement of Carbonaceous Aerosol: Evaluation of the sampling artifacts and inter-comparison of the thermal-optical analysis methods, Atmos. Chem. Phys., 10, 8533-8548, 2010.

Cheng Y., Duan F. K., He K. B., Zheng M., Du Z. Y., Ma Y. L., and Tan J. H.: Intercomparison of thermal-optical methods for the determination of organic and elemental carbon: Influences of aerosol composition and implications, Environ. Sci. Technol., 45, 10117-10123, 2011.

Cheng Y., Duan F. K., He K. B., Du Z. Y., Zheng M., and Ma Y. L.: Intercomparison of thermal-optical method with different temperature protocols: implications from source samples and solvent extraction, Atmos. Environ., 61, 453-462, 2012a.

Cheng Y., Duan F. K., He K. B., Du Z.Y ., Zheng M., and Ma Y. L.: Sampling artifacts of organic and inorganic aerosol: implications for the speciation measurement of particulate matter, Atmos. Environ., 55, 229-233, 2012b.

Section 3. Source apportionment results during the typical summer and winter period.

%	factor 1	factor 2	factor 3	factor 4	factor 5	factor 6
			OC			
summer	0.00	9.80	70.48	0.00	17.49	2.23
winter	0.00	53.97	7.24	0.00	31.19	7.59
			EC			
summer	5.11	0.03	12.41	14.92	42.43	25.10
winter	0.78	0.10	0.77	0.65	45.92	51.77
			WSOC			
summer	9.98	6.22	69.23	2.46	9.45	2.65
winter	3.59	49.01	10.17	0.25	24.10	12.87
			levoglucosan			
summer	1.44	21.18	30.98	11.75	8.15	26.51
winter	0.16	51.69	1.41	0.38	6.44	39.92
			K^+			
summer	57.73	3.94	19.09	11.10	4.16	3.98
winter	24.23	36.19	3.27	1.34	12.39	22.57
			oxalate			
summer	18.88	0.00	59.56	19.10	2.07	0.38
winter	27.54	0.00	35.47	8.01	21.44	7.55

Section 4. Estimating the contributions of the six factors resolved by PMF to the constructed $PM_{2.5}$ mass using the multivariate linear regression.

The contributions of the six factors resolved by PMF to the constructed $PM_{2.5}$ mass were estimated using the multivariate linear regression:

$$\begin{aligned}[Constructed\ PM_{2.5}] = K_1 \times [factor1] + K_2 \times [factor2] + K_3 \times [factor3] + K_4 \times [factor4] \\ + K_5 \times [factor5] + K_6 \times [factor6] + b\end{aligned}$$

where [constructed $PM_{2.5}$] is the constructed $PM_{2.5}$ mass, [factor1] ~ [factor6] is the factor contribution resolved by PMF, K_1 ~ K_6 is the slope of each factor, and b is the constant. In this study, R^2 of the multivariate linear regression was 0.99.

Section 5. Statistical results for the concentrations, ratios, regressions and comparisons included in the present study.

Table S-1 Concentrations of $PM_{2.5}$ components during summer and winter in Beijing. Values of OC, EC and WSOC are presented in μgC/m^3, whereas concentrations of the other species are presented in μg/m^3. Levoglucosan to K^+ and levoglucosan to mannosan ratios of the ambient samples are also shown.

	OC	EC	WSOC	levo.	manno.	K^+	NH_4^+	SO_4^{2-}	NO_3^-	$C_2O_4^{2-}$	Cl^-	Ca^{2+}	Na^+	Mg^{2+}	levo./K^+	levo./manno.
							typical summer									
minimum	5.14	0.92	2.72	0.06	0.00	0.09	0.44	1.44	0.73	0.06	0.04	0.10	n.a.	0.01	0.05	7.44
lower quartile	8.06	1.69	5.10	0.07	0.01	0.38	2.83	7.66	2.20	0.24	0.29	0.29	0.05	0.08	0.10	10.55
median	9.32	2.02	5.88	0.10	0.01	0.68	9.77	19.00	13.72	0.36	0.80	0.45	0.13	0.11	0.16	12.50
upper quartile	11.43	2.54	7.49	0.16	0.01	1.10	16.88	28.64	24.77	0.42	1.37	0.61	0.20	0.13	0.23	14.97
maximum	18.71	4.21	11.95	0.26	0.02	2.60	42.79	70.89	69.74	0.90	6.00	0.80	0.39	0.22	0.69	23.05
							BB summer									
minimum	7.62	1.76	5.05	0.10	0.01	2.33	7.30	23.65	8.56	0.39	0.94	0.24	0.16	0.07	0.04	12.97
lower quartile	15.66	3.14	11.85	0.40	0.02	3.62	14.76	27.05	16.24	0.69	1.25	0.35	0.26	0.12	0.09	18.52
median	20.78	3.95	14.58	0.46	0.02	5.70	15.82	40.55	23.72	0.85	1.78	0.39	0.32	0.14	0.10	21.65
upper quartile	23.99	4.81	16.35	0.65	0.03	7.40	20.17	42.12	29.58	0.89	2.90	0.65	0.37	0.16	0.13	22.98
maximum	38.12	6.15	25.98	2.30	0.12	10.10	27.57	44.92	46.30	1.41	11.27	0.83	0.62	0.21	0.24	56.72
							typical winter									
minimum	5.50	1.77	1.98	0.06	n.a.	0.14	0.73	2.34	0.86	0.03	0.48	0.17	0.06	0.06	0.24	4.39
lower quartile	15.43	4.83	7.00	0.33	0.05	0.80	3.41	5.86	4.89	0.08	3.90	1.19	0.48	0.13	0.40	8.43
median	22.81	7.77	9.17	0.61	0.06	1.02	5.49	9.97	8.63	0.14	5.23	1.45	0.65	0.18	0.48	9.36
upper quartile	29.10	9.44	14.72	0.88	0.10	1.56	10.54	15.95	14.47	0.18	6.82	2.30	0.81	0.22	0.62	9.96
maximum	67.41	28.39	31.08	1.94	0.19	3.16	26.88	45.52	30.69	0.41	14.12	3.15	1.41	0.47	0.82	11.69

							fireworks winter									
minimum	6.05	2.17	2.85	0.08	0.01	0.38	1.15	3.19	1.42	0.05	1.13	0.37	0.21	0.11	0.01	9.52
lower quartile	12.40	3.62	6.16	0.29	0.02	2.23	2.42	8.47	3.37	0.10	3.49	0.66	0.43	0.25	0.10	9.95
median	16.98	4.96	7.91	0.39	0.03	2.70	5.60	12.58	11.18	0.16	5.47	0.79	0.61	0.32	0.15	10.37
upper quartile	38.24	7.82	19.04	0.64	0.06	5.06	16.74	31.32	27.50	0.23	10.28	1.20	1.04	0.44	0.21	11.32
maximum	73.14	21.43	35.76	1.09	0.11	45.76	37.39	70.32	66.65	0.47	28.09	1.65	1.38	1.79	0.32	13.24

Table S-2 Statistical results of the linear regression analysis included in the present study.

Y	X	slope	confidence interval of slope	intercept	confidence interval of intercept	R^2	Note
levo.	K^+	0.05±0.01	0.03~0.08	0.07±0.01	0.05~0.10	0.34	typical summer
levo.	K^+	0.50±0.04	0.43~0.57	0.03±0.05	-0.08~0.13	0.82	typical winter
levo.	manno.	9.93±0.97	7.97~11.90	0.02±0.01	0.00~0.04	0.73	typical summer
levo.	manno.	17.67±1.61	13.85~21.49	0.14±0.08	-0.05~0.33	0.95	BB summer
levo.	manno.	9.41±0.22	8.98~9.85	0.01±0.02	-0.02~0.05	0.97	whole winter

Table S-3 Comparison of levoglucosan to K^+ ratios among different types of biomass: p values of the Independent-Samples T Test ($p < 0.1$ indicates significant difference at a 95% level of confidence, whereas $p > 0.1$ indicates insignificant difference). The Independent-Samples T Test was not performed between EU hardwood and the other types of biomass because few data are available for EU hardwood.

	US hardwood	US softwood	EU hardwood	EU softwood	US needles	US grass	US duff	Asian rice straw
US hardwood		0.306		0.086	0.003	0.019	0.985	0.000
US softwood				0.103	0.007	0.003	0.523	0.001
EU hardwood								
EU softwood					0.063	0.059	0.086	0.054
US needles						0.171	0.028	0.000
US grass							0.003	0.143
US duff								0.008
Asian rice straw								

Table S-4 Comparison of levoglucosan to mannosan ratios among different types of biomass: p values of the Independent-Samples T Test (p＜0.1 indicates significant difference at a 95% level of confidence, whereas p＞0.1 indicates insignificant difference).

	US hardwood	US softwood	EU hardwood	EU softwood	US needles	US grass	US duff	Asian rice straw
US hardwood		0.000	0.455	0.002	0.000	0.357	0.000	0.264
US softwood			0.009	0.448	0.082	0.012	0.000	0.001
EU hardwood				0.008	0.007	0.875	0.005	0.101
EU softwood					0.517	0.010	0.000	0.001
US needles						0.009	0.001	0.001
US grass							0.005	0.071
US duff								0.000
Asian rice straw								

Section 6. The relationship between the levoglucosan to K^+ ratio and the levoglucosan to mannosan ratio: a detailed comparison among different kinds of biomass.

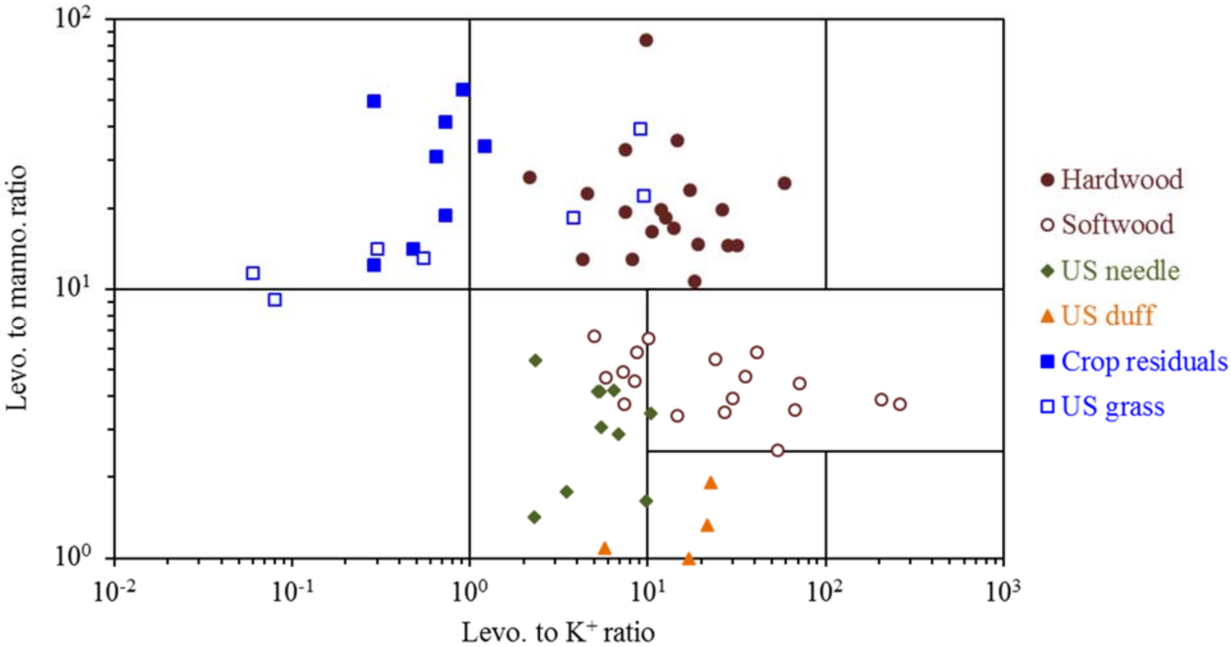

Figure S-1. The levoglucosan to K^+ and the levoglucosan to mannosan ratios of different kinds of biomass.

As shown in Figure S-1, emissions from hardwood burning are characterized by high values of both the levoglucosan to K^+ ratio and the levoglucosan to mannosan ratio which were typically in the range of 1 ~ 100 and 10 ~ 100, respectively. Compared with hardwood, softwood burning resulted in a lower levoglucosan to mannosan ratios (typically in the range of 2.5 ~ 10) whereas the levoglucosan to K^+ ratios (typically in the range of 10 ~ 1000) tended to be higher for softwood burning. US needles had a comparable but wider range of levoglucosan to mannosan ratios (typically between 1 ~ 10) compared with softwood, while they were better separated by the levoglucosan to K^+ ratio which was generally lower for US needles (typically between 1 ~ 10). US duff differed from softwood by its lower levoglucosan to mannosan ratio (typically between 1 ~ 2.5) and differed from US needles by its higher levoglucosan to K^+ ratio which was in general above 10. Crop residuals were distinguished from the other biofuel by their low levoglucosan to K^+ ratios (typically below 1). Therefore, each kind of biomass smoke is characterized by a distinct range of levoglucosan to K^+ and levoglucosan to mannosan ratios.

Some source emissions studies measured only either the levoglucosan to K^+ ratio or the levoglucosan to mannosan ratio. To include results from these studies, the statistical results of the levoglucosan to K^+ ratio and the levoglucosan to mannosan ratio measured in source samples were presented separately in Table S-5 and S-6 (based on the raw data summarized in Section 1). With respect to US grass, though its levoglucosan to K^+ ratio could be as high as 9.50, but the median value of the levoglucosan to K^+ ratio (0.55) was very close to that of Asian rice straw (0.57). In addition, the levoglucosan to mannosan ratios of US grass and Asian rice straw were in the same range (i.e., above 10), and were substantially higher than softwood and US needle. Therefore, in the manuscript, we prefer to classify US grass and Asian rice straw in the same region.

Table S-5. Statistical results of the levoglucosan to K^+ ratio measured in source samples.

	US hardwood	US softwood	EU hardwood	EU softwood	US needle	US grass	US duff	Asian straw
minimum	2.16	4.61		10.12	2.30	0.06	5.72	0.24
lower quartile	7.87	7.30		39.88	3.92	0.19	12.91	0.29
median	**12.65**	**14.61**	**25.69**	**60.16**	**5.42**	**0.55**	**17.06**	**0.57**
upper quartile	19.83	28.69		171.86	6.72	6.42	21.75	0.73
maximum	59.06	71.45		260.87	10.45	9.50	22.73	1.22

Table S-6. Statistical results of the levoglucosan to mannosan ratio measured in source samples.

	US hardwood	US softwood	EU hardwood	EU softwood	US needle	US grass	US duff	Asian straw
minimum	10.67	2.56	11.18	2.53	1.42	9.17	0.99	12.31
lower quartile	15.85	3.45	14.50	3.24	2.94	12.29	1.06	18.80
median	**19.71**	**4.49**	**14.70**	**3.75**	**3.70**	**14.08**	**1.51**	**30.93**
upper quartile	26.48	5.12	22.58	4.29	4.17	20.24	2.02	41.59
maximum	83.43	6.67	34.19	6.54	6.53	39.15	2.43	55.03