Appendix A. Supplementary data

Potassium: A Tracer for Biomass Burning in Beijing?

Jinting Yu, Caiqing Yan, Yue Liu, Xiaoying Li, Tian Zhou, Mei Zheng*

SKL-ESPC and BIC-ESAT, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China

* Corresponding author: mzheng@pku.edu.cn

Uncertainty analysis

The uncertainty analysis for this method is conducted by combining the uncertainties for the elements concentration measurements by the Xact (K, Ca, Pb) and elements ratios (K/Ca, K/Pb), and calculated using the error transfer formula in the following equations.

$$\left(\frac{U_{dust}}{K_{dust}}\right)^2 = \left(\frac{U_{Ca}}{C_{Ca}}\right)^2 + \left(\frac{U_{K/Ca}}{R_{K/Ca}}\right)^2 \quad (1)$$

$$\left(\frac{U_{coal}}{K_{coal}}\right)^2 = \left(\frac{U_{Pb}}{C_{Pb}}\right)^2 + \left(\frac{U_{K/Pb}}{R_{K/Pb}}\right)^2 \quad (2)$$

$$(U_{biomass})^2 = (U_K)^2 + (U_{dust})^2 + (U_{coal})^2 \quad (3)$$

Where $\mathbf{U_{dust}}$**,** $\mathbf{U_{coal}}$ and $\mathbf{U_{biomass}}$ represented the uncertainties of dust, coal and biomass burning source contribution to K;

$\mathbf{K_{dust}}$ and $\mathbf{K_{coal}}$ represented the concentration of dust and coal contribution to K for each samples;

$\mathbf{U_{Ca}}$**,** $\mathbf{U_{Pb}}$ and $\mathbf{U_{K}}$ represented the uncertainties of Ca, Pb and K measured by the Xact, respectively, and use the 5/6 method detection limit of those elements to calculate, MDL: K: 2.36 ng m^{-3}, Ca: 0.90 ng m^{-3} and Pb: 0.22 ng m^{-3};

$\mathbf{C_{Ca}}$ and $\mathbf{C_{Pb}}$ represented the concentration of Ca and Pb for each sample;

$\mathbf{U_{K/Ca}}$ and $\mathbf{U_{K/Pb}}$ represented the uncertainties of the K/Ca and K/Pb ratios used to estimate the dust and coal contribution. $U_{K/Ca}$ was 0.07 and $U_{K/Pb}$ was 3.57.

$\mathbf{R_{K/Ca}}$ and $\mathbf{R_{K/Pb}}$ represented the ratio value of K/Ca and K/Pb used to estimate the dust and coal contribution. For spring, $R_{K/Ca}$ was 0.35 and for all other seasons was 0.15.

Table S1. The percent contribution of K and K^+ to $PM_{2.5}$ as well as the K/K^+ ratio from different source profiles.

	Location_source type	K^+(%)	K(%)	K/K^+	Reference
Cooking	Beijing_domestic kitchen	0.04	0.11	2.97	Zhang *et al.*, 2017
	Shanxi_street food	0.83	1.76	2.12	Zhang *et al.*, 2016
	Shanxi_hot pot	0.41	0.50	1.22	
	Shanxi_barbecue	1.69	2.32	1.37	
	Shanxi_restaurant	0.19	0.65	3.42	
Biomass Burning	Shannxi_Wheat straw	4.33	7.19	1.66	KLACP, 2017
	Shandong_Wheat straw	3.79	4.44	1.17	
	Shannxi_Maize straw	2.72	5.47	2.01	
	Shannxi_Maize straw	2.34	3.99	1.71	
	Wheat straw	5.49	7.35	1.34	Tian, 2016
	Rice straw	2.47	3.68	1.49	
Coal Combustion	Shanxi_coal power plant	0.25	1.73	7.03	Wang, 2016
	Shanxi_coal power plant	0.18	1.90	10.68	
	Shanxi_coal power plant	0.16	1.75	10.92	
	Beijing_coal-fired boilers	2.69	2.87	1.06	Wang *et al.*, 2009
	Beijing_coal-fired boilers	2.20	3.48	1.59	
	Beijing_coal-fired boilers	1.00	1.04	1.04	KLACP, 2017
	Shannxi_coal-fired boilers	0.61	0.86	1.41	
	Shannxi_residetial coal	0.40	0.76	1.90	
	Shannxi_residetial coal	0.83	1.95	2.35	
Dust	Beijing_road dust	0.17	0.94	5.53	KLACP, 2017
	Beijing_road dust	0.11	0.91	8.27	
	Beijing_construction dust	0.19	1.18	6.21	
	Beijing_construction dust	0.15	0.59	3.93	
	Beijing_road dust	0.17	1.09	6.46	Shen *et al.*, 2016
	Beijing_construction dust	0.23	1.20	5.17	
	Tianjin_road dust	0.18	1.03	5.75	
	Tianjin_construction dust	0.17	1.22	7.32	

Table S2. Pearson correlations between $PM_{2.5}$ and trace metals across the four seasons.

	$PM_{2.5}$_Autumn		$PM_{2.5}$_Winter		$PM_{2.5}$_Spring		$PM_{2.5}$_Summer	
	r	n	r	n	r	n	r	n
K	0.92**	592	0.83**	648	0.90**	622	0.77**	695
Pb	0.91**	592	0.85**	648	0.90**	622	0.68**	695
Zn	0.88**	592	0.89**	648	0.88**	622	0.68**	695
Fe	0.81**	592	0.85**	648	0.66**	622	0.68**	695
Mn	0.78**	592	0.83**	648	0.85**	622	0.67**	695
Cu	0.86**	592	0.79**	648	0.80**	622	0.62**	695
Ca	0.58**	592	0.68**	648	0.23**	622	0.23**	695
Ba	0.85**	592	0.82**	648	0.41**	622	0.51**	695
As	0.85**	391	0.78**	553	0.79**	512	0.67**	479
Cr	0.52**	464	0.78**	590	0.51**	524	0.36**	575
Se	0.92**	448	0.85**	633	0.92**	566	0.81**	600
Ni	0.65**	556	0.66**	634	0.78**	600	0.47**	639

**Correlation is significant at the 0.01 level.
"n" indicates the number of samples used in correlation analyses.
"r" indicates the correlation coefficient.

Table S3. Ratios (Mean ± S.D.) of K/Pb and K/As under different $PM_{2.5}$ concentration levels across the four seasons.

Ratio	Season	$PM_{2.5}<10^{th}$ percentiles	$10^{th}<PM_{2.5}<90^{th}$ percentiles	$PM_{2.5}>90^{th}$ percentiles
K/Pb	Spring	31.83±16.03	26.77±19.38	15.41±2.79
	Summer	37.10±29.88	24.98±18.24	14.27±3.88
	Autumn	39.06±24.18	29.71±15.60	17.71±2.48
	Winter	13.65±6.42	14.73±4.66	15.40±3.97
K/As	Spring	223.74±64.93	126.54±138.63	61.05±11.83
	Summer	127.47±43.37	162.13±96.16	77.07±29.12
	Autumn	162.87±85.46	143.21±94.43	53.17±17.11
	Winter	176.62±74.94	105.81±66.75	66.63±10.56

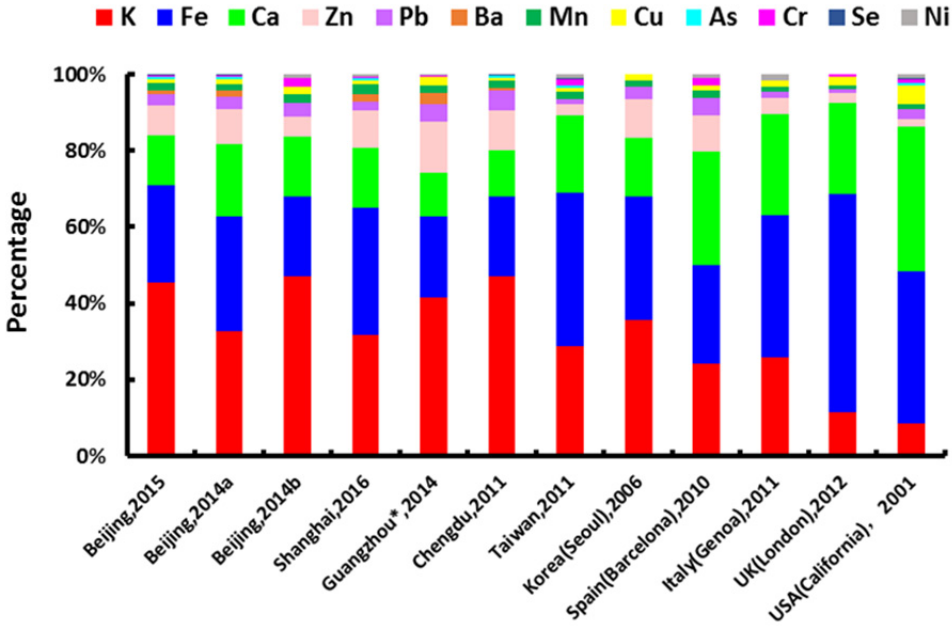

Fig. S1. Relative contributions of different metals to the select metals measured in this study ~~total metals measured~~ in different cities. Metals include K or K^+. The references for the different cities are as follows: Beijing, 2015 (this study), Beijing, 2014a (Gao *et al.*, 2016), Beijing, 2014b (Li *et al.*, 2017), Shanghai, 2016 (Chang *et al.*, 2017), Guangzhou, 2014 (Tao *et al.*, 2017), Chengdu, 2011 (Tao *et al.*, 2014), Taiwan, 2011 (Gugamsetty *et al.*, 2012), Korea (Seoul), 2006 (Heo *et al.*, 2009), Spain(Barcelona), 2010 (Dall'Osto *et al.*, 2013), Italy (Genoa), 2011 (Salameh *et al.*, 2015), UK (London), 2012 (Visser *et al.*, 2015), and USA (California), 2001 (Na *et al.*, 2009). All studies used K, except the study in Guangzhou, which used K^+.

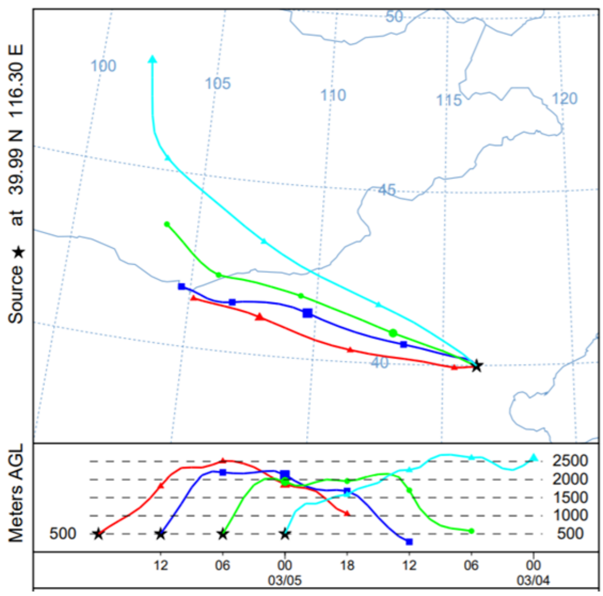

Fig. S2. 24-h backward trajectories for dust episode on March 4th and March 5th, 2016 from the HYSPLIT model.

Reference

Chang, Y.H., Huang, K., Deng, C.R., Zou, Z., Liu, S.D. and Zhang, Y.L. (2017). First long-term and near real-time measurement of atmospheric trace elements in Shanghai, China. *Atmos. Chem. Phys. Discuss.*doi:10.5194/acp-2017-613

Dall'Osto, M., Querol, X., Amato, F., Karanasiou, A., Lucarelli, F., Nava, S., Calzolai, G. and Chiari, M. (2013). Hourly elemental concentrations in PM2.5 aerosols sampled simultaneously at urban background and road site during SAPUSS–diurnal variations and PMF receptor modelling. *Atmos. Chem. Phys.* 13: 4375–4392.

Gao, J., Peng, X., Chen, G., Xu, J., Shi, G.L., Zhang, Y.C. and Feng, Y.C. (2016). Insights into the chemical characterization and sources of PM2.5 in Beijing at a 1-h time resolution. Sci. Total Environ. 542:162-171.

Gugamsetty, B., Wei, H., Liu, C.H., Awasthi, A., Hsu, S.C., Tsai, C.J., Roam, G.D., Wu, Y.C. and Chen, C.F. (2012). Source Characterization and Apportionment of PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ by Using Positive Matrix Factorization. *Aerosol Air Qual. Res.*12:476-491.

Heo, J.B., Hopke, P.K. and Yi, S.M. (2009). Source apportionment of PM2.5 in Seoul, Korea. *Atmos. Chem. Phys.* 9:4957–4971

KLACP (Key Laboratory of Aerosol Chemistry and Physics, Chinese Academy of Sciences). (2017). PM2.5 main source profiles database in China, http://www.sourceprofile.org.cn/.

Li, Y.Y., Chang, M., Ding, S.S., Wang, S.W., Ni, D. and Hu, H.T. (2017). Monitoring and source apportionment of trace elements in PM2.5: Implications for local air quality management. *J. Environ. Manage.*196:16-25.

Na, K. and Cocker, D.R. (2009). Characterization and source identification of trace elements in PM2.5 from Mira Loma, Southern California. *Atmos.Res.*93:793-800.

Salameh, D., Detournay, A., Pey, J., Pérez, N., Liguori, F., Saraga, D., Bove, M.C., Brotto, P., Cassola, F., Massabò, D., Latella, A., Pillon, S., Formenton, G., Patti, S., Armengaud, A., Piga, D., Jaffrezo, J.L., Bartzis, J., Tolis, E., Prati, P., Querol, X., Wortham, E. and Marchand, N. (2015). PM2.5 chemical composition in five European Mediterranean cities: A 1-year study. *Atmos.Res.*155:102-117.

Shen, Z.X., Sun, J. Cao, J.J., Zhang, L.M., Zhang, Q., Lei, Y.L., Cao, J.J., Huang, R.J., Liu, S.X., Huang, Y.H., Zhu, C.S., Xu, H.M., Zheng, C.L., Liu, P.P. and Xue, Z.G. (2016). Chemical profiles of urban fugitive dust PM2.5 samples in Northern Chinese cities. *Sci. Total Environ.* 569: 619-626.

Tao, J., Gao, J., Zhang, L., Zhang, R., Che, H., Zhang, Z., Jing, J., Cao, J. and Hsu, S.C. (2014). PM2.5 pollution in a megacity of southwest China: source apportionment and implication. *Atmos. Chem. Phys*.14:8679-8699

Tao, J., Zhang, L.M., Cao, J.J., Zhong, L.J., Chen, D.S., Yang, Y.H., Chen, D.H., Chen, L.G., Zhang, Z.S., Wu, Y.F., Xia, Y.J., Ye, S.Q. and Zhang, R.J. (2017). Source apportionment of PM2.5 at urban and suburban areas of the Pearl River Delta region, south China - With emphasis on ship emissions. *Sci. Total Environ*.

574:1559-1570.
Tian, J. (2016). Laboratory study on emission characteristics of PM2.5 from crop residue burning and residential coal combustion in China (in Chinese). The university of Chinese Academy of Science.
Visser, S., Slowik, J.G., Furger, M., Zotter, P., Bukowiecki, N., Dressler, R., Flechsig, U., Appel, K., Green, D.C., Tremper, A.H., Young, D.E., Williams, P.I., Allan, J.D., Herndon, S.C., Williams, L.R., Mohr, C., Xu, l., Ng, N.L., Detournay, A., Barlow, J.F., Halios, C.H., Fleming, Z.L., Baltensperger, U. and Prévôt, A.S.H. (2015) Kerb and urban increment of highly time-resolved trace elements in PM10, PM2.5 and PM1.0 winter aerosol in London during ClearfLo 2012. *Atmos. Chem. Phys.* 15: 2367-2386.
Wang, S.X., Zhao, X.J., Li, X.H., Wei, W. and Hao, J.M. (2009). Emission characteristics of fine particles from grate firing boilers (in Chinese). *Environ. Sci.* 30: 963–968.
Wang, Y.X. (2016). Study on source profiles and risk assessment of heavy metals in power plants coal dust. Taiyuan University of Technology.
Zhang, N., Han, B., He, F., Xu, J., Zhao, R.J., Zhang, Y.J. and Bai, Z.P. (2017). Chemical characteristic of PM2.5 emission and inhalational carcinogenic risk of domestic Chinese cooking. *Environ. Pollut.* 227:24-30.
Zhang, T., Peng, L., Li, Y.H., Liu, H.L., Wang, Y.X. and Wang, Y. (2016). Chemical characteristics of PM2.5 emitted from cooking fumes (in Chinese). *Research of Environment Science.* 29:183-191.