Supplementary Material

Appendix 1.	The calculation method of SOC.
Table S1	Reference information of sampling period, site and analysis method
Fig. S1	Comparison between average concentration of OC and coal consumption from 2006-2014.
Fig. S2	Comparison between annual average $PM_{2.5}$ concentration and meteorology parameters. (northerly wind include N, NNE, NE, ENE, W, WNW, NW and NNW; southerly wind include E, ESE, SE, SSE, S, SSW, SW and WSW).
Fig. S3	SOR and annual average relative humidity during 2000-2015.

Appendix 1.

The secondary organic carbon (SOC) in 2011-2015 was further estimated using the EC tracer method as follows:

$$POC = aEC + b \quad (1)$$

$$SOC = OC - POC = OC - (aEC + b) \quad (2)$$

where a and b are the slope and intercept, respectively. aEC represents the primary OC associated with combustion source (coal burning and traffic) (Liu *et al*., 2016). The intercept b is the non-combustion source contribution to the primary OC (e.g., cooking operations and biogenic sources) and sampling artifacts (Cabada *et al*., 2004; Lin *et al*., 2009). This method supposes a and b are constant at a certain location during the season. The main uncertainty of this method is from the estimation of a and b and the primary OC/EC ratio is crucial to the validity of the method. Two steps were applied to reduce the uncertainty (Cabada *et al*., 2004). First, data points with rain and storms were eliminated because significant changes of OC/EC ratio (e.g., the preferential remove of SOC and aged particles, the increased impact of the locally produced ones) were caused by extreme weather. Second, selected the OC and EC concentrations with lowest probability of SOC production. Lowest 5-20% OC/EC ratios in each season were used to approximate the primary OC/EC ratio (Cao *et al*., 2007). Thus, slope a and intercept b were calculated by four to five samples with the lowest OC/EC ratios by least-square regression in each season during 2011-2015 (Zhao *et al*., 2013c). Most of the correlation coefficient of the least-square regression was greater than 0.9. Slope a was found higher in winter (3.5) and lower in summer (1.3), the same as Zhao *et al*. (2013c). The large number of coal combustion in winter increase the contribution of combustion source. The intercept “b” calculated in this study (1.39) was similar to the results of Ji *et al*. (2016) and Liu *et al*. (2016), and was found similar seasonal variation with “a”.

Table S1. Reference information of sampling period, site and analysis method

Reference	Sampling period	Site	Analysis method
He et al., 2001	1999.7-2000.9	THU[a] , CGZ[b]	XRF[1], IC[2], TOR[3]
He et al., 2011	1999.9-2008.8	THU, CGZ	XRF, IC, TOR
Yang et al., 2005	1999.7-2000.6	THU, CGZ	TOR, XRF
Zheng et al., 2005	2000 1, 4, 7, 10	PKU[c], DS EPB[d]	IC, XRF, TOM[4], GC-MS[5]
Song et al., 2006	2000 1, 4, 7, 10	PKU, DS EPB	IC, XRF, TOM
Duan et al., 2006	2001.8-2002.9	THU, CGZ	XRF, IC, TP[6]
Zheng et al., 2015	2001-2012	USE[e]	-
Sun et al., 2004	2002, 2003 (i)	BNU[f]	IC, ICP-AES[7], C/H/N[8]
Wang et al., 2014b	2002.7- 2003.7	SJFCEP[g]	GC-FID[9], IC
Xu et al., 2007	2003.1-5 2003.8-2004.1	BDA[h], CMA[i]	NAA[10], PIXE[11], IC, AAS[12]
Yu et al., 2009	2004 1, 4, 7, 10	PKU, OSC[j]	TOM, GC-MS
Zhao et al., 2009	2005.1-2007.12	BL[k]	-
Yang et al., 2011a	2005.1-2008.12	THU	TOM
Lin et al., 2009	2005.11-2006.10	PKU	TP
Wang et al., 2015b	2005.1-2010.12	IAP[l]	-
Zhou et al., 2012a	2006 (ii)	IAP	IC, TOM
Chen et al., 2014	2007.9-2011.1	CRAES[m]	HR-ICP-MS[13]
Hu et al., 2015	2008.1-2009.12	CMA	IC, TOM
Song et al., 2012	2008.7-2009.12	4th [n]	WD-XRF[14], IC, TOR
Zhao et al., 2013b	2009 (iii)	BMS [o]	IC, ICP-AES, TOM
Martini et al., 2015	2008.4-2014.7	USE	-
Zhang et al., 2013	2009.4-2010.1	PKU	IC, ICP-MS
Zhao et al., 2013a	2010.1-12	Urban	-
Yu et al., 2013	2010.1-12	BNU	PIXE
Wang et al., 2015a	2012.6-2013.4	BHU[p]	IC, XRF, TOM
Wang et al., 2016b	2012.8-2013.7	CGZ	IC, ICP-AES
Wang et al., 2016a	2013.12-2015.4	CRAES	-
Chen et al., 2015	2014.5-2015.4	Urban	-

[a] THU: Tsinghua University

[b] CGZ:Chegongzhuang

[c] PKU: Peking University

[d] DS EPB: Dongsi Environmental Protection Board

[e] USE: U.S. Embassy

[f] BNU: Beijing Normal University

[g] SJFCEP: Sino–Japan Friendship Centre for Environmental Protection

[h] BDA: Beijing Dancing Academy

[i] CMA: China Meteorological Administration

[j] OSC: Olympic Sports Center
[k] BL: Baolian
[l] IAP: Institute of Atmospheric Physics
[m] CRAES: Chinese Research Academy of Environmental Science
[n] 4th: North 4th Ring
[o] BMS: Beijing Meteorological Service
[p] BHU: Beihang University
[1] XRF: X-ray fluorescence
[2] IC: ion chromatography
[3] TOR: thermal/optical reflectance method
[4] TOM: thermal/optical method
[5] GC-MS: gas chromatography/mass spectrometry
[6] TP: temperature program
[7] ICP-AES: inductively coupled plasma atomic emission spectroscopy
[8] C/H/N: C/H/N elemental analyzer
[9] GC-FID: gas chromatography - flame ionization detection
[10] NAA: neutron activation analysis
[11] PIXE: proton induced X-ray emission
[12] AAS: atomic absorption spectroscopy
[13] HR-ICP-MS: high-resolution inductively coupled plasmamass spectrometer
[14] WD-XRF: wavelength dispersive X-ray fluorescence spectrometer
(i) June 18th to July 20th, December 1th to 31th in both 2002 and 2003
(ii) March 16th to April 6th, July 19th to August 31th, October 23th to November 13th, and December 6th to December 29th
(iii) April 6th to May 1th, July 9th to August 4th, October 11th to November 4th in 2009 and January 14th to February 8th in 2010

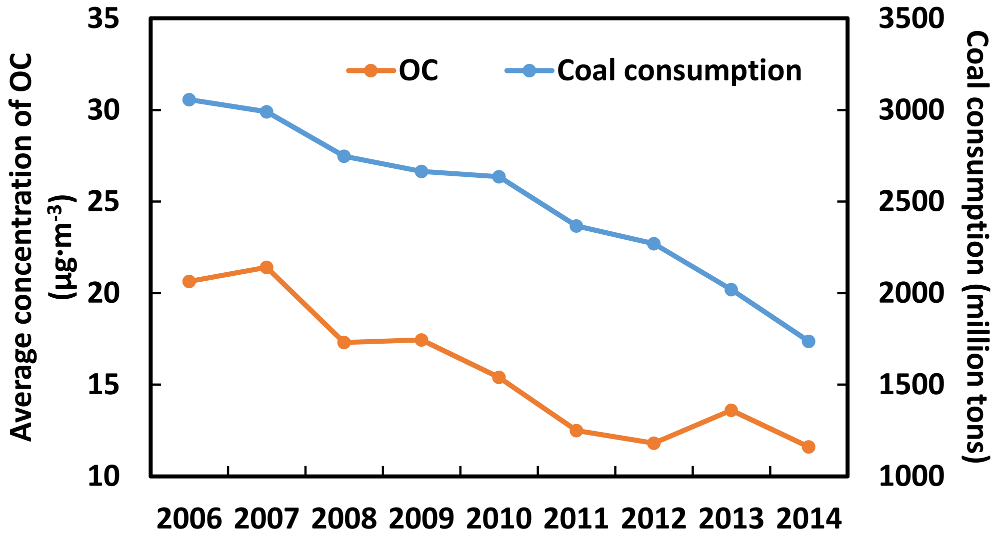

Fig S1. Comparison between average concentration of OC and coal consumption from 2006-2014.

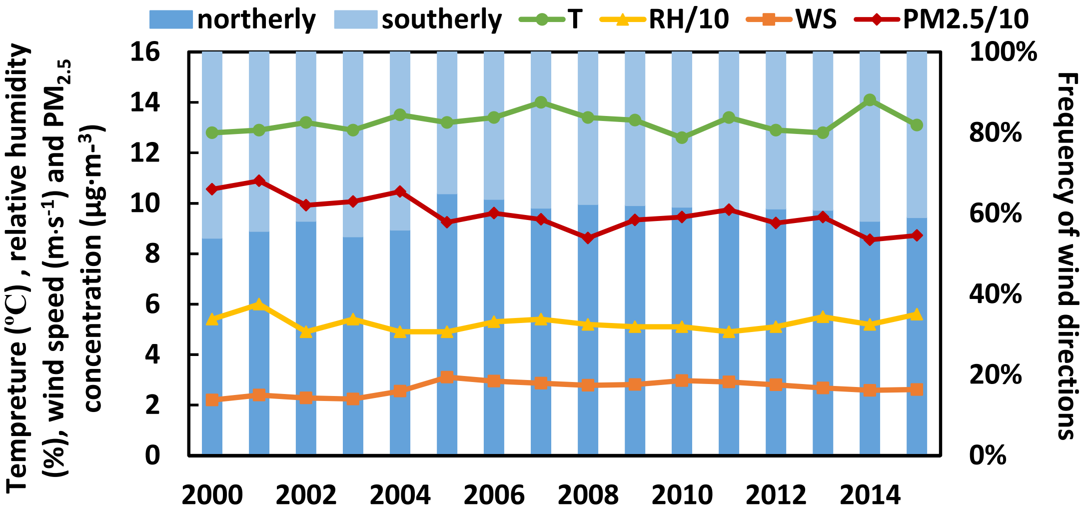

Fig. S2. Comparison between annual average $PM_{2.5}$ concentration and meteorology parameters. (northerly wind include N, NNE, NE, ENE, W, WNW, NW and NNW; southerly wind include E, ESE, SE, SSE, S, SSW, SW and WSW).

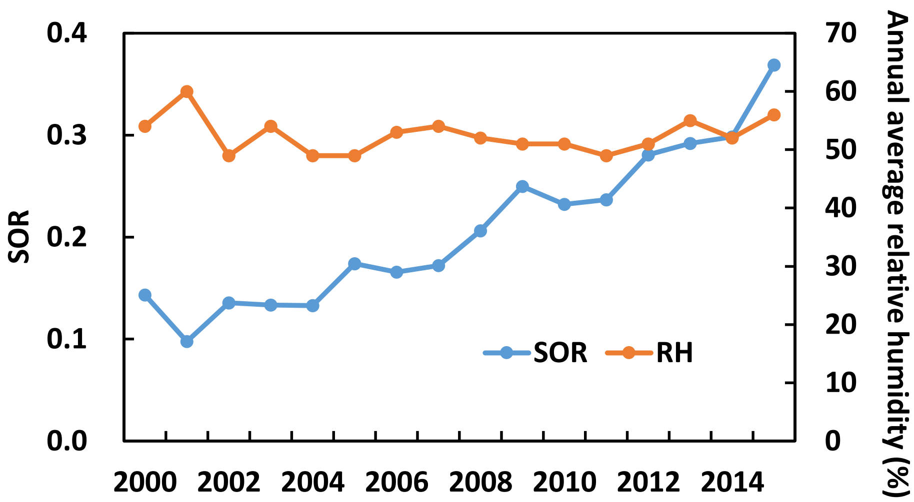

Fig. S3. SOR and annual average relative humidity during 2000-2015.

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