



Supplement of

Source apportionment of carbonaceous aerosols in Xi'an, China: insights from a full year of measurements of radiocarbon and the stable isotope $^{13}{\rm C}$

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S1 Sample selection for radiocarbon (¹⁴C) measurements

There are 58 PM_{2.5} samples in total, with 13 collected in spring, 15 in summer, 12 in autumn, and 18 in winter. Six samples with varying PM_{2.5} mass and carbonaceous aerosols loading were selected per season for ¹⁴C analysis. Air mass back trajectories (for identifying the probable sources and transport pathways of air pollutions) are also considered when selecting samples for ¹⁴C analysis. 72h air mass back trajectories starting 150 m above ground level at 2:00 UTC (10:00 a.m., local standard time) were calculated using NOAA HYSPLIT trajectory model. The best situation is that the back trajectories were similar between days with high PM_{2.5} loading and low-to-medium PM_{2.5} loading, in which case, the influence of air pollution transport to the sampling site could be minimized. Back trajectories of selected samples are presented in Fig. S2.

S2 Measurement of source markers (levoglucosan, hopanes, picene and water-soluble potassium)

Organic markers including levoglucosan, picene and hopanes were quantified using the Gas chromatography-mass spectrometry (GC/MS) instrumentation. Filter pieces were extracted with a mixture of dichloromethane and methanol (2:1, v/v) under ultrasonication. The extracts were concentrated using a rotary evaporator in vacuum and then blown down to dryness using a pure nitrogen stream. After reaction with N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) at 70 °C for 3 h, the derivatives were determined using a GC/MS technique. GC/MS analysis of the derivatized fraction was performed using an Agilent 7890A GC coupled with an Agilent 5975 CMSD. The GC separation was carried out on a HP-5MS fused silica capillary column with the GC oven temperature programmed from 50 °C (2 min) to 120 °C at 15 °C min⁻¹ and then to 300 °C at 5 °C min⁻¹ with a final isothermal hold at 300 °C for 16 min. The sample was injected in a splitless mode at an injector temperature of 280 °C, and scanned from 50 to 650 Daltons using electron impact (EI) mode at 70 eV. GC/MS response factors were determined using authentic standards. We use the sum of measured hopanes (Σ hopanes) in this study, including $17\alpha(H)$,22,29,30-Trisnorhopane, $17\alpha(H)$,21 $\beta(H)$ -30- $17\beta(H), 21\alpha(H)-30$ -Norhopane, $17\alpha(H), 21\beta(H)$ -Hopane, Norhopane, $17\alpha(H), 21\alpha(H)$ -Hopane, $17\beta(H), 21\alpha(H)$ -Hopane, $17\alpha(H), 21\beta(H)-(22S)$ -Homohopane, and $17\alpha(H), 21\beta(H)-(22R)$ -Homohopane.

Water-soluble potassium (K⁺) was measured in water extracts using Ion Chromatography (Dionex 600, Thermal Scientific-Dionex, Sunnyvale, CA, USA). IonPac CS12A column was used for the separation of cations. 20mM methanesulfonic acid with a flow rate of 1 mL min⁻¹ was utilized as eluent for cation separation. The minimum detection limit for K⁺ was 0.001 μ g mL⁻¹. Details of these measurements are described in Li et al. (2016a) and Zhang et al. (2011).

S3 Determination of modern and fossil contamination for radiocarbon measurement

 $F^{14}C$ of aerosols samples was corrected for contamination that occurred during graphitization and AMS measurement. For AMS measurements, samples are usually analysed together with varying amounts of reference material covering the range of sample mass. Two such materials with known ¹⁴C content are used: the oxalic acid OXII calibration material ($F^{14}C = 1.3406$) and a ¹⁴C-free CO₂ gas ($F^{14}C = 0$).

Contamination during the graphitization and AMS measurement results into the differences between measured and nominal $F^{14}C$ values. The magnitude of these deviations can be used to quantify the contamination with fossil carbon ($F^{14}C_F = 0$) and modern carbon ($F^{14}C_M = 1$), which in turn are used for correcting the sample values (de Rooij et al., 2010).

The contamination with fossil carbon and modern carbon is quantified using isotope mass balance (Dusek et al., 2014):

$$F^{14}C_{m} \cdot M_{m} = F^{14}C_{st} \cdot M_{st} + F^{14}C_{F} \cdot M_{F} + F^{14}C_{M} \cdot M_{M}.$$
(S1)

 M_m and M_{st} stand for the experimentally determined mass and the mass of reference materials either the oxalic acid OXII calibration material ($F^{14}C = 1.3406$) or a ¹⁴C-free CO₂ gas ($F^{14}C = 0$) with a unit of µgC, respectively. $F^{14}C_m$ and $F^{14}C_{st}$ represent the experimentally determined $F^{14}C$ measured by AMS and nominal $F^{14}C$ of reference materials (Table S9).

The relationships among all masses are described as Eq. (S2):

$$M_{\rm m} = M_{\rm st} + M_{\rm F} + M_{\rm M}, \tag{S2}$$

where M_M is calculated using Eq. (S1) by substituting $F^{14}C_{st} = 0$ for a ¹⁴C-free CO₂ gas as:

$$\mathbf{M}_{\mathrm{M}} = \mathbf{F}^{14} \mathbf{C}_{\mathrm{m}} \cdot \mathbf{M}_{\mathrm{m}}. \tag{S3}$$

Substitute $F^{14}C_{st} = 1.3406$ for OXII and the derived M_M from Eq. (S3), M_F is derived by combining Eq. (S1) and Eq. (S2) as:

$$M_{\rm F} = ((1.3406 - F^{14}C_{\rm m}) \cdot M_{\rm m} - (1.3406 - 1) \cdot M_{\rm M})/1.3406.$$
(S4)

 M_M and M_F are calculated by applying Eq. (S3) and Eq. (S4), and they are mass dependent. The modern carbon contamination (M_M) is between 0.35 and 0.50 µg C, and the fossil carbon contamination (M_F) is typically around 2 µg C for a sample bigger than 100 µgC.

S4 Primary OC/EC ratios from biomass burning (r_{bb}), coal combustion (r_{coal}), and liquid fossil fuel combustion ($r_{liq.fossil}$)

There is considerable variability in the published OC/EC ratios for biomass burning (Fig. S16), coal combustion (Fig. S17) and liquid fossil fuel combustion (e.g., vehicle emissions; Table S10). OC/EC ratios differ due to variabilities in experimental factors, such as fuel types and properties, combustion conditions (e.g., smoldering vs. flaming), sampling and analysis methods (e.g., different protocols for OCEC measurements) etc.

When selecting OC/EC ratios for each source, we applied the following rules: first, we prioritize localized measurements of fresh emissions and estimations specific to China; second, OCEC measured by IMPROVE_A (Chow et al., 2007), the same protocol applied in this study, have higher priorities than those measured by other protocols. This is because different protocols (e.g., IMPROVE_A, IMPROVE and NIOSH) can lead to differences in OC/EC ratios up to over 3 times (Chow et al., 2001). The difference in OC/EC ratios between IMPROVE and IMPROVE_A can be up to a factor of 2 (Chow et al., 2007); third, for sources with limited data, average of the available data is used.

Biomass burning emissions are mixtures of emissions from crop residues open burning, crop residues

burning in household stove, and wood burning in household stove etc. Higher OC/EC ratios are reported for crop residues open burning than those reported for similar fuels burned in household stove (Ni et al., 2015), perhaps due to a more complete combustion of household biofuels leading to higher EC emission. As it is difficult to estimate the distribution among different biomass burning subtypes, here we take OC/EC ratios from emission inventories, where major types of biomass burning are included. OC and EC emission amounts from previously reported emission inventories are summarized and their ratios are presented in Fig. S16. OC/EC ratios ranged from 3 to 7, with the mean of 4.4. This range covers the OC/EC ratios from fresh emissions of wood combustion (e.g., 4.67 for wood burning in rural China by Shen et al. (2015)) and crop residues burning (e.g., 7 for mixture of wheat straw, rice straw and corn stalk by Han et al. (2016)). We took $r_{bb} = 5 (3-7)$ to account for the variabilities of biomass burning emissions. A bit lower central value of 4.5 was used in previous ¹⁴C-based source apportionments (Zhang et al., 2014, 2015b).

OC/EC ratios for fresh emissions from <u>coal combustion</u> are summarized in Fig. S17. A relatively wide range of OC/EC ratios is found, which partially can be explained by different protocol applied to OC/EC measurements (Chow et al., 2001, 2004; Han et al., 2016). We took OC/EC ratios in the literature measured by IMPROVE_A protocol when available, to be consistent with our OCEC measurements. 1.4 ± 1.3 and 6.3 ± 1.3 (average ± 1 standard deviation) are used as OC/EC ratios from bituminous and anthracite coal combustion, respectively. They are quantified from typical used coals in residential sector in China (Tian et al., 2017), and measured by IMPROVE_A protocol (Chow et al., 2007). The selected OC/EC ratios of 1.4 ± 1.3 and 6.3 ± 1.3 for bituminous and anthracite coal, respectively, overlap with most of the data in the literature (Fig. S17). The final OC/EC ratio for coal combustion (r_{coal}) depends on the share of bituminous coal and anthracite coal. Bituminous and anthracite coal are apportioned 80% and 20% respectively, according to raw coal production data (Chen et al., 2005; Zhi et al., 2008), leading to $\underline{r_{coal}} = 2.38 \pm 0.44$ derived from Monte Carlo Simulation with assumption of triangular distribution. This ratio is similar to 2.25 derived from OC and EC emission amounts from emission inventories of coal combustion for 2000, and 2.26 for the year 2005 estimated by Zhi et al. (2008).

<u>Vehicle emissions</u> can be influenced by vehicle type, fuel quality, speed of the vehicle, as well as the features of the road (He et al., 2008; Cheng et al., 2010; Cui et al., 2016). Literature searches were conducted (Table S10) to establish OC/EC ratios for vehicle emissions. Due to the limitation of published data, the lower/upper bonds were estimated as the mean of all lower/upper bounds from different datasets. The mean was then calculated as the average of the lower and upper bounds. The established $r_{liq.fossil}$ ranged from 0.69 to 1.01 with the mean of 0.85.



Figure S1. Temporal variability of OC and EC mass concentrations in $PM_{2.5}$ in Xi'an, China, during July 2008 to June 2009 (n=58). Twenty-four samples were selected for ¹⁴C analysis and highlighted in light green. Details on sample selection are presented in Supplemental S1.



Figure S2. Three-day backward-in-time air mass trajectory analysis of selected samples for radiocarbon measurements.



Figure S3. Temporal variation of levoglucosan to EC mass ratios (levo/EC), Σ hopanes to EC ratios (Σ hopanes/EC), picene to EC ratios (picene/EC) for samples selected for radiocarbon measurements. Details of measurements are in Supplemental S2.



Figure S4. Fire counts (red points) monitored by MODIS in different seasons during the sampling period (<u>https://firms.modaps.eosdis.nasa.gov/firemap/</u>). The sampling site is Xi'an. Xi'an is located in the Guanzhong Plain, one of the major agricultural production areas.



Figure S5. Correlation between $F^{14}C_{(EC)}$ and levoglucosan/EC ratios, K⁺/EC ratios in different seasons (red: autumn; dark green: spring; blue: summer; winter: black). One data point with extremely high K⁺ concentration on Chinese New Year is removed.



Figure S6. The δ^{13} C variability for EC from burning C4 plants. In this study, δ^{13} C for corn stalk is used as it is the dominant C4 plant in Xi'an and its surrounding areas (Guanzhong Plain). The range used as δ^{13} C of burning corn stalk is indicated as dashed vertical lines, and the mean is shown by a solid vertical line. Sources: ¹Chen et al. (2012), ²Guo et al. (2016), ³Liu et al. (2014), ⁴Kawashima and Haneishi (2012), ⁵Martinelli et al. (2002), ⁶Das et al. (2010).



Figure S7. MCMC3-derived posterior probability density functions (PDF) of the relative source contributions of C3 plants (denoted as C3), coal and liquid fossil fuel combustion (vehicle) to EC in different seasons, calculated using the Bayesian Markov chain Monte Carlo approach.



Figure S8. MCMC4-derived posterior probability density functions (PDF) of the relative source contributions of liquid fossil fuel combustion (vehicle), coal and biomass burning (C3 and C4 plants, denoted as biomass) to EC in different seasons (a), calculated using the Bayesian Markov chain Monte Carlo approach. The PDF of the relative source contributions of biomass burning (a) is a posteriori combination of PDF for C3 plants and C4 plants, as shown in panel (b).



Figure S9. Comparison between the MCMC-derived fraction of biomass burning EC (f_{bb} derived from MCMC4) and that obtained from radiocarbon data (¹⁴C-based $f_{bb}(EC)$). Average and one standard deviation is shown for $f_{bb}(EC)$, median with interquartile range is shown for f_{bb} . f_{bb} derived from MCMC3 is also very similar to $f_{bb}(EC)$.



Figure S10. Sources of EC in different seasons. Results from the $F^{14}C$ and $\delta^{13}C$ based Bayesian source apportionment calculations of EC. The numbers in the bars represent the median contribution of liquid fossil fuel, coal and biomass burning. (a) results from the MCMC3 model, including C3 plants as biomass, coal and liquid fossil fuel; (b) Impact of C4 plants burning on EC source apportionment is tested by including C4 biomass into the calculations (MCMC4). For MCMC4, the PDF for C3 and C4 plants is combined and named as biomass burning. Bars filled with green colour indicate the relative contribution of biomass burning, including C4 plants (light green) and C3 plants (dark green). In winter, the sample taken on Chinese New Year eve (25 January 2009) was excluded.



Figure S11. MCMC4-derived source contributions to EC for each data point computed using the Bayesian Markov chain Monte Carlo approach. (a). biomass burning from C3 plants; (b). biomass burning from C4 plants; (c). liquid fossil fuel combustion; (d). coal combustion. Range of 95 % credible intervals (Bayesian analogue of confidence intervals) and interquartile range (25th-75th percentile) from the computed probability density functions (PDF) and shown in black and green error bars, respectively. To better compare results with MCMC3, we did a posteriori combination of PDF for C3 biomass (a) and C4 biomass (b) and named the combined PDF as biomass burning (e).



Figure S12. Estimated primary OC based on MCMC3 results. (a) measured OC concentrations (blue line and diamond symbols) with observational uncertainties (vertical bar) and estimated OC mass (OC_{pri,e}, circle and triangular symbols) from apportioned EC and OC/EC ratios for different sources (Eq. (10)). (b) ¹⁴C-based fraction of non-fossil OC ($f_{nf}(OC)$) and modelled non-fossil fraction in OC_{pri,e} ($f_{bb}(OC_{pri,e})$) derived from Eq. (11). Interquartile range (25th-75th percentile) of the median OC_{pri,e} and $f_{bb}(OC_{pri,e})$ are shown in purple (A), red (B) and green (C) vertical bars. "A" and "B" denote different OC/EC ratios applied to primary biomass burning emissions (r_{bb}): A. $r_{bb} = 5$ (3–7, minimum-maximum), B. $r_{bb} = 4$ (3–5). "C" denotes 80 % $r_{liq,fossil}$ applied in summer with $r_{bb} = 5$. $f_{nf}(OC)$ uncertainties are indicated but are too small to be visible.



Figure S13. Observed and estimated OC concentrations. Modelled $OC_{e,min}$ is the sum of $OC_{pri,e}$ and $OC_{o,nf}$. $OC_{o,nf}$ accounts for the differences between $f_{nf}(OC)$ and $f_{bb}(OC_{pri,e})$, with an unrealistic assumption of no secondary fossil OC, leading to minimum addition to $OC_{pri,e}$. Coral area shows the POC_{bb,e} and $OC_{o,nf}$, green area the POC_{coal,e} and blue area the POC_{liq.fossil,e}. Estimation is based on MCMC3 results for EC source apportionment and primary OC/EC ratios corresponding to case (A) in Fig. S12.



Figure S14. Probability density functions (PDF) of the relative source contributions of coal combustion to EC in winter in the year 2008/2009 (this study, shown in grey; this is also shown in Fig. S8) and 2012/2013 by Wang et al. (2016), shown in yellow.



Figure S15. Probability density functions (PDF) of the relative source contributions of vehicle emissions to EC in winter in the year 2008/2009 (this study, shown in grey; this is also shown in Fig. S8) and 2012/2013 by Wang et al. (2016), shown in yellow.



Figure S16. OC/EC ratios estimated from OC and EC emission amounts from biomass burning emission inventories specific to China. y-axis on the right side indicates the year of estimation. The range applied in OC estimation (Sect. 4.4 in main text) is shown by dashed vertical lines, and the mean is indicated by a full vertical line. Data sources: Streets et al. (2003); Yan et al. (2006); Zhang et al. (2006, 2009); Cao et al. (2006, 2011a), Qin and Xie (2011), Zhang et al. (2013), Zhou et al. (2017).



Figure S17. Literature reported OC/EC ratios for combustion of bituminous coal and anthracite coal. Boxplots show the median (thick line across the box), interquartile range (25th-75th percentile, vertical ends of the box). Outliers are shown as triangles. Blue dots (averages) with error bars (one standard deviation) represent OC/EC ratios measured by IMPROVE_A protocol reported by Tian et al. (2017). Data sources: Chen et al. (2005, 2006, 2015), Zhang et al., (2008, 2012), Zhi et al. (2008), Shen et al. (2010, 2015), Li et al. (2016b).

Sources	δ^{13} C of emissions from sources (ranges)	Source signatures of $\delta^{13}C$ used in the source apportionment calculations of EC (mean \pm standard deviation)	Reference
Corn stalk	-19.30 ‰ to -13.6 ‰ ^a	$-16.4 \pm 1.4 \%^{a}$	(Martinelli et al., 2002; Das et al., 2010; Chen et al., 2012; Kawashima and Haneishi, 2012; Liu et al., 2014; Guo et al., 2016)
C3 plants (wood, wheat straw, etc.)	-35 ‰ to -24 ‰	-26.7 ± 1.8 ‰	Andersson et al. (2015) and references therein
coal	-25 ‰ to -21 ‰	-23.4 ± 1.3 ‰	Andersson et al. (2015) and references therein
liquid fuel (e.g., gasoline, diesel, and oil)	-28 ‰ to -24 ‰	-25.5 ± 1.3 ‰	Andersson et al. (2015) and references therein

Table S1. Range of δ^{13} C values for each source reported in previous studies.

^a δ^{13} C source signature for EC from burning corn stalk (C4 plant) of -16.4 ± 1.4 ‰ (mean ± standard deviation) is applied in MCMC4 calculations. In this study, δ^{13} C for corn stalk is used as it is the dominant C4 plant in Xi'an and its surrounding areas (Sun et al., 2017; Zhu et al., 2017), with little sugarcane and other C4 plants. See details on selection of δ^{13} C signature for C4 plants in the study area (corn stalk) in Sect.4.3.1 and Fig. S6.

	PM _{2.5} (µg m ⁻³)	OC (µg m ⁻³)	EC (µg m ⁻³)
Spring (n=13)	124.0 ± 40.4 (55.9–193.4) ^a	14.4 ± 9.6 <i>(3.3–33.8)</i>	5.7 ± 2.3 (2.0-8.8)
Summer (n=15)	83.0 ± 30.7 <i>(31.8–139.2)</i>	12.7 ± 4.5 <i>(4.0–20.6)</i>	6.3 ± 2.0 (2.7–10.0)
Autumn (n=12)	125.1 ± 69.3 <i>(41.0–212.6)</i>	22.2 ± 13.6 <i>(3.6–34.2)</i>	8.4 ± 2.9 <i>(3.5–11.3)</i>
Winter (n=18)	213.4 ± 91.8 <i>(73.1–408.5)</i>	39.0 ±17.8 (10.8–67.0)	9.1 ± 3.1 <i>(5.6–16.3)</i>
Annual	142.0 ± 82.4 <i>(31.8–408.5)</i>	21.5 ± 16.6 <i>(3.3–67.0)</i>	7.6 ± 3.0 (2.0–16.3)

Table S2. Mass concentrations of $PM_{2.5}$, OC and EC in Xi'an, China from July 2008 to June 2009.

 $^{\rm a}\, average \pm$ standard deviation, the numbers in parentheses are the range of each dataset.

Date	F ¹⁴ C (OC)	$F^{14}C \ ({\rm EC})$	$\delta^{13}Coc$	$\delta^{13}C_{EC}$	Season
7/17/2008ª	0.466 ± 0.010	0.178 ± 0.003	-26.80	-26.50	summer
7/23/2008	0.489 ± 0.008	0.164 ± 0.003	-25.94	-26.33	summer
8/4/2008	0.546 ± 0.007	0.153 ± 0.002	-25.86	-26.16	summer
8/11/2008	0.512 ± 0.008	0.141 ± 0.003	-25.21	-25.53	summer
9/3/2008	0.549 ± 0.006	0.129 ± 0.002	-25.94	-26.23	autumn
10/3/2008	0.581 ± 0.006	0.166 ± 0.002	-24.55	-25.51	autumn
10/16/2008	0.659 ± 0.007	0.188 ± 0.002	-23.70	-24.31	autumn
10/21/2008	0.610 ± 0.005	0.301 ± 0.003	-24.51	-24.92	autumn
11/2/2008	0.651 ± 0.006	0.172 ± 0.002	-24.94	-25.10	autumn
11/14/2008	0.579 ± 0.007	0.200 ± 0.004	-25.48	-24.79	autumn
11/26/2008	0.671 ± 0.009	0.245 ± 0.004	-24.71	-22.93	winter
12/20/2008	0.696 ± 0.008	0.225 ± 0.002	-24.06	-22.81	winter
1/1/2009	0.693 ± 0.007	0.317 ± 0.004	-23.23	-23.12	winter
1/25/2009	0.745 ± 0.005	0.505 ± 0.008	-23.39	-23.07	winter
2/6/2009	0.671 ± 0.007	0.318 ± 0.005	-23.92	-23.72	winter
3/5/2009	0.572 ± 0.006	0.183 ± 0.003	-25.44	-23.53	winter
3/17/2009	0.545 ± 0.004	0.177 ± 0.002	-25.72	-26.03	spring
3/29/2009	0.547 ± 0.006	0.153 ± 0.002	-26.91	-25.38	spring
4/16/2009	0.545 ± 0.007	0.166 ± 0.003	-27.42	-25.05	spring
4/22/2009	0.535 ± 0.006	0.175 ± 0.004	-26.33	-25.27	spring
4/28/2009	0.330 ± 0.021	0.175 ± 0.005	-26.41	-25.33	spring
5/4/2009	0.544 ± 0.004	0.180 ± 0.003	-26.66	-25.35	spring
6/9/2009	0.549 ± 0.006	0.132 ± 0.003	-24.24	-25.37	summer
6/21/2009	0.489 ± 0.006	0.124 ± 0.002	-26.30	-25.73	summer
summer ^b	0.509 ± 0.033	0.149 ± 0.020	-25.7 ± 0.9	-25.9 ± 0.5	
autumn	0.605 ± 0.044	0.193 ± 0.058	$\textbf{-24.9}\pm0.8$	-25.1 ± 0.7	
winter	0.675 ± 0.057	0.299 ± 0.114	$\textbf{-24.1}\pm0.8$	-23.2 ± 0.4	
spring	0.508 ± 0.087	0.171 ± 0.010	-26.6 ± 0.6	-25.4 ± 0.4	

Table S3. Average fraction modern ($F^{14}C$) and stable carbon signature ($\delta^{13}C$, ‰) of OC and EC for selected samples.

 a Daily $F^{14}\mathrm{C}$ values are given in average \pm measurement uncertainty;

 b Seasonal averaged $F^{14}C$ and $\delta^{13}C$ values are given in average \pm standard deviation.

 OC_{nf} $f_{nf}(OC)$ $f_{\text{fossil}}(\text{OC})$ $f_{bb}(EC)$ $f_{\text{fossil}}(\text{EC})$ Date OC_{fossil} EC_{bb} **EC**_{fossil} Season 7/17/2008 3.53 ± 0.50 4.48 ± 0.63 1.13 ± 0.26 5.87 ± 1.29 0.440 ± 0.010 0.560 ± 0.010 0.162 ± 0.004 0.838 ± 0.004 summer 7/23/2008 4.70 ± 0.75 5.49 ± 0.89 1.20 ± 0.24 6.84 ± 1.41 0.461 ± 0.009 0.539 ± 0.009 0.149 ± 0.004 0.851 ± 0.004 summer 8/4/2008 5.56 \pm 0.85 5.22 ± 0.80 1.01 ± 0.21 6.21 ± 1.26 0.515 ± 0.008 0.485 ± 0.008 0.139 ± 0.003 0.861 ± 0.003 summer 8/11/2008 6.16 ± 0.93 6.59 ± 0.99 0.96 ± 0.21 6.51 ± 1.41 0.483 ± 0.009 0.517 ± 0.009 0.128 ± 0.003 0.872 ± 0.003 summer 9/3/2008 6.54 ± 1.05 6.09 ± 0.98 1.17 ± 0.18 8.81 ± 1.32 0.518 ± 0.007 0.482 ± 0.007 0.117 ± 0.003 0.883 ± 0.003 autumn 10/3/2008 13.65 ± 1.80 0.849 ± 0.003 11.26 ± 1.50 1.54 ± 0.31 8.64 ± 1.75 0.548 ± 0.007 0.452 ± 0.007 0.151 ± 0.003 autumn 10/16/2008 21.23 ± 3.11 12.95 ± 1.86 1.94 ± 0.32 9.40 ± 1.57 0.622 ± 0.008 0.378 ± 0.008 0.171 ± 0.004 0.829 ± 0.004 autumn 12.82 ± 1.78 10/21/2008 9.41 ± 1.32 2.65 ± 0.44 7.03 ± 1.13 0.576 ± 0.007 0.424 ± 0.007 0.274 ± 0.006 0.726 ± 0.006 autumn 20.42 ± 3.22 11/2/2008 12.82 ± 2.03 1.60 ± 0.22 8.62 ± 1.20 0.614 ± 0.008 0.386 ± 0.008 0.156 ± 0.003 0.844 ± 0.003 autumn 11/14/2008 3.83 ± 0.60 3.16 ± 0.50 0.89 ± 0.18 4.00 ± 0.78 0.546 ± 0.008 0.454 ± 0.008 0.182 ± 0.005 0.818 ± 0.005 autumn 14.49 ± 1.80 8.41 ± 1.05 11/26/2008 1.91 ± 0.36 6.66 ± 1.25 0.634 ± 0.010 0.366 ± 0.010 0.223 ± 0.006 0.777 ± 0.006 winter 12/20/2008 36.16 ± 4.43 18.83 ± 2.37 1.69 ± 0.43 6.56 ± 1.69 0.657 ± 0.009 0.343 ± 0.009 0.204 ± 0.004 0.796 ± 0.004 winter 1/1/2009 38.59 ± 4.92 20.39 ± 2.65 4.69 ± 0.71 11.62 ± 1.72 0.654 ± 0.008 0.346 ± 0.008 0.288 ± 0.007 0.712 ± 0.007 winter 1/25/2009 32.79 ± 4.19 4.34 ± 1.34 0.703 ± 0.007 0.297 ± 0.007 0.459 ± 0.011 13.86 ± 1.78 5.10 ± 1.58 0.541 ± 0.011 winter 2/6/2009 17.71 ± 2.74 10.27 ± 1.61 2.68 ± 0.50 6.58 ± 1.19 0.633 ± 0.008 0.367 ± 0.008 0.289 ± 0.007 0.711 ± 0.007 winter 3/5/2009 9.36 ± 1.15 7.98 ± 0.98 0.99 ± 0.22 4.97 ± 1.08 0.540 ± 0.007 0.460 ± 0.007 0.166 ± 0.004 0.834 ± 0.004 winter 3/17/2009 17.38 ± 2.58 16.39 ± 2.47 1.41 ± 0.27 7.33 ± 1.37 0.514 ± 0.006 0.486 ± 0.006 0.161 ± 0.003 0.839 ± 0.003 spring 3/29/2009 13.05 ± 1.77 12.21 ± 1.67 1.22 ± 0.19 7.56 ± 1.15 0.517 ± 0.007 0.483 ± 0.007 0.139 ± 0.003 0.861 ± 0.003 spring 4/16/2009 6.33 ± 0.80 0.87 ± 0.19 4.87 ± 1.07 5.98 ± 0.76 0.515 ± 0.008 0.485 ± 0.008 0.151 ± 0.004 0.849 ± 0.004 spring 4/22/2009 3.84 ± 0.68 3.77 ± 0.68 0.84 ± 0.17 4.43 ± 0.89 0.505 ± 0.007 0.495 ± 0.007 0.159 ± 0.005 0.841 ± 0.005 spring 4/28/2009 2.28 ± 0.35 5.03 ± 0.72 0.58 ± 0.12 3.08 ± 0.63 0.311 ± 0.019 0.689 ± 0.019 0.159 ± 0.005 0.841 ± 0.005 spring 5/4/2009 8.86 ± 1.12 8.41 ± 1.08 0.513 ± 0.006 0.487 ± 0.006 0.837 ± 0.004 1.30 ± 0.25 6.65 ± 1.30 0.163 ± 0.004 spring 6/9/2009 7.07 ± 0.98 6.57 ± 0.92 0.91 ± 0.17 6.72 ± 1.23 0.518 ± 0.007 0.482 ± 0.007 0.120 ± 0.004 0.880 ± 0.004 summer 3.81 ± 0.66 0.113 ± 0.003 0.887 ± 0.003 6/21/2009 4.47 ± 0.78 0.52 ± 0.13 4.07 ± 1.02 0.462 ± 0.006 0.538 ± 0.006 summer

Table S4. Average OC and EC concentrations from non-fossil sources (OC_{nf} , EC_{bb}) and fossil sources (OC_{fossil} , EC_{fossil}), relative non-fossil sources contribution to OC and EC ($f_{nf}(OC)$, $f_{bb}(EC)$), and relative fossil sources contribution to OC and EC ($f_{fossil}(OC)$, $f_{fossil}(EC)$).

Season	OC _{nf}	OC _{fossil}	EC _{bb}	EC _{fossil}	$f_{\rm nf}({ m OC})$	$f_{\rm fossil}({ m OC})$	$f_{\rm bb}({ m EC})$	$f_{\text{fossil}}(\text{EC})$
summer	$5.14 \pm 1.38^{\rm a}$	5.47 ± 0.95	0.95 ± 0.24	6.03 ± 1.02	0.480 ± 0.032	0.520 ± 0.032	0.135 ± 0.018	0.865 ± 0.018
	(3.53 ~ 7.07)	(4.47 ~ 6.59)	(0.52 ~ 1.20)	(4.07 ~ 6.84)	(0.440 ~ 0.518)	(0.482 ~ 0.560)	(0.113 ~ 0.162)	(0.838 ~ 0.887)
autumn	13.08 ± 7.06	9.28 ± 3.94	1.63 ± 0.62	7.75 ± 2.00	0.571 ± 0.041	0.429 ± 0.041	0.175 ± 0.053	0.825 ± 0.053
	(3.83 ~ 21.23)	(3.16 ~ 12.95)	(0.89 ~ 2.65)	(4.00 ~ 9.40)	(0.518 ~ 0.622)	(0.378 ~ 0.482)	(0.117 ~ 0.274)	(0.726 ~ 0.883)
winter ^b	23.26 ± 13.25	13.18 ± 5.96	2.39 ± 1.42	7.28 ± 2.53	0.624 ± 0.048	0.376 ± 0.048	0.234 ± 0.054	0.766 ± 0.054
	(9.36 ~ 38.59)	(7.98 ~ 20.39)	(0.99 ~ 4.69)	(4.97 ~ 11.62)	(0.540 ~ 0.657)	(0.343 ~ 0.460)	(0.166 ~ 0.289)	(0.711 ~ 0.834)
spring	8.62 ± 5.74	8.63 ± 4.83	1.04 ± 0.32	5.65 ± 1.80	0.479 ± 0.082	0.521 ± 0.082	0.155 ± 0.009	0.845 ± 0.009
	(2.28 ~ 17.38)	(3.77 ~ 16.39)	(0.58 ~ 1.41)	(3.08 ~ 7.56)	(0.311 ~ 0.517)	(0.483 ~ 0.689)	(0.139 ~ 0.163)	(0.837 ~ 0.861)
overall ^b	12.06 ± 9.81	8.96 ± 4.79	1.46 ± 0.90	6.65 ± 1.96	0.535 ± 0.080	0.465 ± 0.080	0.172 ± 0.051	0.828 ± 0.051
	(2.28 ~ 38.59)	(3.16~20.39)	(0.52 ~ 4.69)	(3.08 ~ 11.62)	(0.311 ~ 0.657)	(0.343 ~ 0.689)	(0.113 ~ 0.289)	(0.711 ~ 0.887)

Table S5. OC and EC concentrations from non-fossil sources (OC_{nf}, EC_{bb}) and fossil sources (OC_{fossil}, EC_{fossil}), relative non-fossil sources contribution to OC and EC ($f_{nf}(OC)$, $f_{bb}(EC)$), and relative fossil sources contribution to OC and EC ($f_{fossil}(OC)$, $f_{fossil}(EC)$) in different seasons.

^a data is given in average ± standard deviation, minimum and maximum are shown in parentheses

^b the sample taken on Chinese New Year eve (25 January 2009) was excluded.

Location	l	Site type	Sampling period	Seasons	PM fraction	δ^{13} Coc (‰)	$\delta^{13}\mathrm{C}_\mathrm{EC}$ (‰)	References
	Beijing, China	urban	Jan, 2013	winter	PM _{2.5}	$\textbf{-24.26} \pm 0.29$		Yan et al. (2017)
	Beijing, China	urban	Feb, 2010	winter	PM _{2.5}		-25.1 to -24.2	Chen et al. (2013)
North	North China Plain	urban	Jan, 2013	winter	PM _{2.5}		-24.3 to -23.3	Andersson et al. (2015)
China	7 cities in North China	urban	Jan 6–20, 2003	winter	PM _{2.5}	-25.54 to -23.08	-25.02 to -23.27	Cao et al. (2011b)
	Beijing, China	urban	June, 2013	summer	PM _{2.5}	-26.74 ± 0.65		Yan et al. (2017)
	7 cities in North China	urban	June 3–July 30, 2003	summer	PM _{2.5}	-26.90 to -26.33	-26.62 to -25.27	Cao et al. (2011b)
	Hong Kong	urban	Nov 2000–Feb 2001	winter	PM _{2.5}	$\textbf{-26.9}\pm0.6$	$\textbf{-25.6} \pm 0.1$	Ho et al. (2006)
	Shanghai, China	urban	Jan, 2010	winter	PM _{2.5}		-25.8 to -24.7	Chen et al. (2013)
	Xiamen, China	urban	Dec, 2009	winter	PM _{2.5}		-25.3 to -24.9	Chen et al. (2013)
	Pearl River Delta	urban	Jan, 2013	winter	PM _{2.5}		-26.7 to -25.7	Andersson et al. (2015)
South China	Yangtze River Delta	urban	Jan, 2013	winter	PM _{2.5}		-27.7 to -25	Andersson et al. (2015)
	7 cities in South China	urban	Jan 6–20, 2003	winter	PM _{2.5}	-26.62 to -25.79	-26.10 to -25.33	Cao et al. (2011b)
	Shanghai, China	urban	Sept 1-20, 2009	Autumn	PM _{2.5}	$\textbf{-24.5}\pm0.8$	-25.1 ± 0.6	Cao et al. (2013)
	Hong Kong	urban	June–August, 2001	summer	PM _{2.5}	-26.9 ± 0.5	$\textbf{-25.6}\pm0.1$	Ho et al. (2006)
	7 cities in South China	urban	June 3–July 30, 2003	summer	PM _{2.5}	-26.74 to -25.29	-26.63 to -25.41	Cao et al. (2011b)
North	Xi'an China	urban		winter	PM _{2.5}	$\textbf{-24.1}\pm0.8$	$\textbf{-23.2}\pm0.4$	This study
China				autumn	PM _{2.5}	$\textbf{-24.9}\pm0.8$	-25.1 ± 0.7	
				summer	PM _{2.5}	-25.7 ± 0.9	-25.9 ± 0.5	
				spring	PM _{2.5}	$\textbf{-26.6} \pm 0.6$	$\textbf{-25.4}\pm0.3$	
				annual	PM _{2.5}	-25.3 ± 1.2	-24.9 ± 1.1	

Table S6. Stable carbon isotopes for aerosols in China.

	Seasons	summer	autumn	winter ^b	spring	annual ^b
Biomass burning	median	0.136	0.177	0.221	0.156	0.173
(C3 plants)	25th-75th percentile	(0.129–0.142)	(0.16–0.197)	(0.196–0.242)	(0.153–0.159)	(0.166–0.18)
	95% credible intervals	(0.113–0.159)	(0.117–0.245)	(0.106–0.288)	(0.145–0.167)	(0.15–0.196)
coal combustion	median	0.147	0.323	0.644	0.251	0.328
	25 th -75 th percentile	(0.086–0.23)	(0.221–0.436)	(0.534–0.709)	(0.167–0.346)	(0.25–0.403)
	95% credible intervals	(0.025–0.494)	(0.061–0.673)	(0.165–0.805)	(0.055–0.56)	(0.117–0.557)
liquid fossil	median	0.717	0.497	0.136	0.594	0.499
	25th-75th percentile	(0.633–0.778)	(0.383–0.607)	(0.076–0.245)	(0.498–0.677)	(0.423–0.578)
	95% credible intervals	(0.365–0.842)	(0.147–0.774)	(0.022–0.61)	(0.282–0.79)	(0.269–0.712)

Table S7. MCMC3 results^a from the F¹⁴C- and δ^{13} C-based Bayesian Source Apportionment Calculations of EC (Median, interquartile range (25th-75th percentile), and 95 % Credible Intervals).

^a Three main source categories were differentiated using this technique: C3 plants (e.g., wood and crop residue), coal and liquid fossil fuel (e.g., oil, diesel, and gasoline) combustion.

^b the sample taken on Chinese New Year eve (25 January 2009) was excluded.

Table S8. Contribution of C3 and C4 plants burning to EC from the MCMC4 results^a (Median, interquartile range (25th-75th percentile), and 95% Credible Intervals).

	Seasons	summer	autumn	winter ^c	spring	annual ^c
Biomass burning ^b	median	0.099	0.123	0.088	0.113	0.123
from C3 plants	25 th -75 th percentile	(0.080-0.114)	(0.094–0.149)	(0.058–0.122)	(0.092–0.129)	(0.105–0.139)
	95% credible intervals	(0.037–0.137)	(0.041–0.199)	(0.019–0.196)	(0.045–0.149)	(0.07–0.165)
Biomass burning	median	0.035	0.051	0.152	0.042	0.05
from C4 plants	25 th -75 th percentile	(0.022–0.054)	(0.033–0.074)	(0.119–0.182)	(0.027–0.063)	(0.034–0.066)
	95% credible intervals	(0.007–0.097)	(0.012–0.128)	(0.052–0.240)	(0.010-0.11)	(0.013–0.097)

^aResults from the four-sources (C3 biomass, C4 biomass, coal and liquid fossil fuel) MCMC4 model.

^bContribution of biomass burning is shown in Table 2 and done by a posteriori combination of PDF for C3 plants and that for C4 plants.

°Sample taken from Chinese New Year eve (25 January 2009) was excluded.

Standards		nominal F ¹⁴ C	measured F ¹⁴ C	measured mass (M _m ,
			$(F^{14}C_m)$	μgC)
Combustion	OXII	1.3406	1.327 ± 0.022	65
processes ^a	OXII	1.3406	1.321 ± 0.012	117
	anthracite	0	0.020 ± 0.001	51
	anthracite	0	0.002 ± 0.001	75
	anthracite	0	0.004 ± 0.001	219
	anthracite	0	0.005 ± 0.001	254
Graphitization and	¹⁴ C-free CO ₂ gas	0	0.008 ± 0.001	42
¹⁴ C measurements ^b	¹⁴ C-free CO ₂ gas	0	0.004 ± 0.000	81
	¹⁴ C-free CO ₂ gas	0	0.005 ± 0.000	91
	¹⁴ C-free CO ₂ gas	0	0.004 ± 0.000	123
	¹⁴ C-free CO ₂ gas	0	0.003 ± 0.000	162
	¹⁴ C-free CO ₂ gas	0	0.002 ± 0.000	186
	¹⁴ C-free CO ₂ gas	0	0.003 ± 0.000	287
	OXII	1.3406	1.268 ± 0.013	45
	OXII	1.3406	1.270 ± 0.012	81
	OXII	1.3406	1.280 ± 0.011	96
	OXII	1.3406	1.305 ± 0.010	128
	OXII	1.3406	1.337 ± 0.010	162
	OXII	1.3406	1.306 ± 0.006	214
	OXII	1.3406	1.311 ± 0.005	321

Table S9. Measured $F^{14}C$ values and masses of the standards with their nominal $F^{14}C$ values.

^a For combustion processes, two sets of standard material: the oxalic acid HOxII and anthracite with known ¹⁴C contents ($F^{14}C = 1.3406$ and $F^{14}C = 0$, respectively) were combusted using the aerosol combustion system and used for quality control;

^b Varying amounts of reference materials covering the range of sample mass are graphitized and analyzed together with samples in the same wheel of AMS, to correct for contamination during graphitization and AMS measurement.

 Table S10. Published OC/EC ratios for vehicle emissions specific to China.

OC/EC ratios	Sampling site	Measurement protocol	References
0.6 ± 0.2	Shing Mun tunnel (Hongkong, China)	IMPROVE ^a	Cheng et al. (2010)
0.8 ± 0.1	roadside sites (Hongkong, China)	IMPROVE ^a	Cheng et al. (2010)
$0.86^{*} (0.48 - 1.45)^{**}$	Wuzushan tunnel (Yantai, China)	IMPROVE ^a	Cui et al. (2016)
1.03*(0.77-1.35)**	Zhujiang tunnel (Guangzhou, China)	NIOSH ^b	Dai et al. (2015)
$\begin{array}{c} 0.49 \pm 0.04 \\ \left(0.44 0.57 \right)^{**} \end{array}$	Zhujiang tunnel (Guangzhou, China)	NIOSH ^b	He et al. (2008)
0.57	Zhujiang tunnel (Guangzhou, China)	NIOSH ^b	Huang et al. (2006)
1.45	Zhujiang tunnel (Guangzhou, China)	NIOSH ^b	Zhang et al. (2015a)

*represents averaged values;
** represents the range of values;
^a Chow et al. (2004);
^b Birch and Cary, (1996).

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