Supporting Information for "**Emission characteristics for polycyclic aromatic hydrocarbons from solid fuels burned in domestic stoves in rural China**"

Guofeng SHEN[†,‡], Shu TAO[*,†], Yuanchen CHEN[†], Yanyan ZHANG[†], Siye WEI[†], Miao XUE, [†] Bin WANG[†], Rong WANG[†], Yan LY[†], Wei LI[†], Huizhong SHEN[†], Ye HUANG[†], Han CHEN[†]

[†] *Laboratory of Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing, 100871, China*

[‡] *Key Laboratory of Environmental Engineering, Jiangsu Academy of Environmental Sciences, Nanjing 210036, China*

Corresponding author phone and fax: 0086-10-62751938, email: taos@pku.edu.cn

S1. Combustion experiment and fuel/stove properties

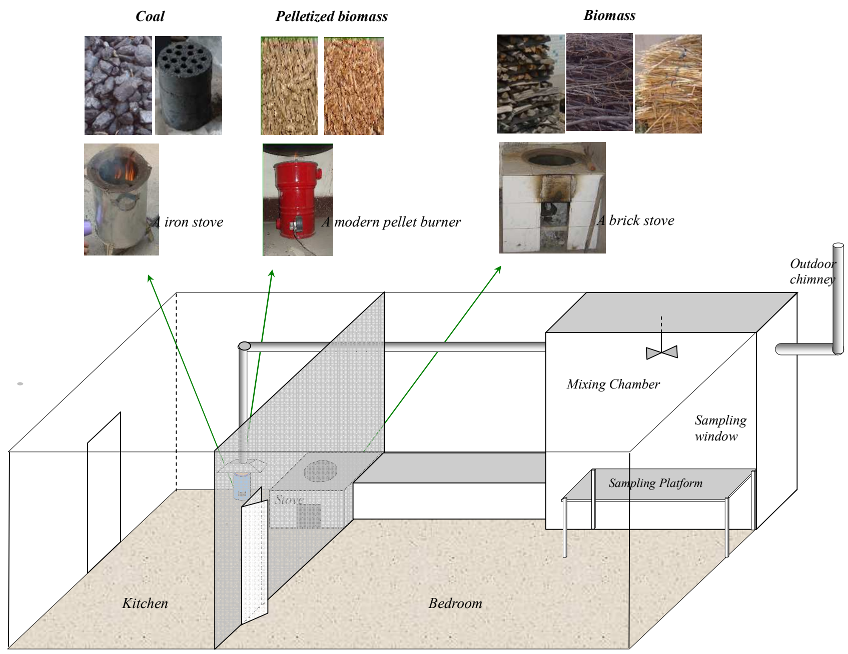

Figure S1 Fuel-stove combinations and the experimental set up in this study

Table S1. Fuel-stove combinations and test numbers of combustion experiments in which emissions of PAHs are measured. Fuel moisture and modified combustion efficiency (MCE) are also collected and used in PAH EF data analysis.

Fuel type	Stove	Moisture, %	MCE, %	n
Crop residue	a brick stove with one pot and a chimney[1]	1.4-7.9	85.8-97.2	25
Wood	the same stove as that used for crop residue	5.3-42	87.8-96.5	76
Coal	a movable iron stove without chimney[1]	0.72-9.2[3]	82.8-94.1	24
Pellet	a modern pellet burner without chimney[2]	5.6-5.8%	95.1-99.8	23

[1] The picture of the so-called brick stove can be found in Shen *et al.,* (2010).

[2] The picture of the modern pellet burner can be found in Shen *et al.,* (2012).

[3] In addition to moisture, volatile matter contents of coals are also collected. Coal volatile matter content is in the range of 3.6-32%.

Chinese rural households typically use brick stoves for biomass combustion and portable iron stoves for coal. **Figure S1** shows the experimental setup in this study. The information about the fuels, the stoves, and burning conditions is summarized in the **Table S1.** For crop residues, in addition to the nine previously reported residues with moisture between 1.4% and 3.9%, three more including stalks of kaoliang (*Sorghum bicolor var. nervosum*), pea (*Pisum sativum*), and maize (*Zea mays*) with moisture between 6.8% and 7.9% (wet basis) were measured and included in this study (not reported previously). The combustion experiments were conducted following the common practice burning solid fuels for daily cooking by rural residents. Pre-weighed fuels were inserted into the stove chamber in batches. The smoke emitted entered into a mixing chamber where all online measurements and sampling were conducted.

CO_2 and CO were measured using on-line non-dispersive infrared detectors (GXH-3051, Junfang Technical Institute of Physical and Chemistry, China). Gaseous and particulate phase PAHs were collected using polyurethane foam plugs (PUFs) and quartz fiber filters (QFFs), respectively. The PUF was Soxhlet extracted using 150 mL dichloromethane for eight h, and particle-loaded QFFs were extracted by a mixture of hexane and acetone using a microwave accelerated system (CEM, Mars Xpress, USA). The extracts were purified using a silica/alumina gel column, concentrated to about one mL, spiked with deuterated internal standards and then analyzed by a gas chromatograph equipped with a mass spectrometer (GC 6890 and MS 5973, Agilent) using a HP-5MS capillary column. The oven temperature was held at 50 °C for one min, increased to 150 °C at a rate of 10 °C/min, to 240 °C at 3 °C/min, and then to 280 °C held for another 20 min. All solvents were from Beijing Reagent, China and re-distillated and checked for PAHs blank before use. The silica gel and alumina were baked at 450 °C for 6 h, activated at 300 °C for 12 h, and deactivated with deionized water (3 %, w/w) prior to use. The anhydrous sodium sulfate was baked at 450 °C for 8 h. All glassware was cleaned in an ultrasonic cleaner and baked at 500 °C for at least 10 h.

EFs were calculated based on the carbon mass balance method which assumed that all the emitted carbon formed CO_2, CO, total hydrocarbon species in gaseous phase and carbon in particulate phase. PAH EFs can be calculated by multiplying CO_2 EFs and the mass emission ratios of PAHs to CO_2. The advantage of the carbon mass balance method is that it's not necessary to collect all emitted species and the sampling site in the plume is adjustable.

S2. Means and standard deviations of EFs of PAHs for various fuels

Table S2. Means of emission factors (mg/kg) of individual PAH, total 15 PAHs and BaPeq for different fuels. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC).

	CR	CP	WP	FW	BW	AB	BB	AC	BC
ACY	3.29	1.31	0.62	1.27	6.98	12.11	6.71	23.88	43.16
ACE	3.39	0.09	0.08	0.10	0.19	2.86	4.00	9.53	8.80
FLO	4.46	0.30	0.30	0.35	1.47	6.50	25.45	18.98	27.29
PHE	7.60	1.75	2.14	2.01	7.83	7.76	42.45	18.70	61.55
ANT	1.06	0.23	0.32	0.28	1.32	2.09	12.74	3.63	24.02
FLA	3.47	0.84	0.65	0.96	8.10	3.36	16.48	3.43	33.91
PYR	3.02	0.73	0.52	0.82	7.66	3.13	18.10	2.70	28.77
BaA	0.61	0.20	0.10	0.23	2.51	0.62	18.18	1.35	23.42
CHR	0.64	0.30	0.18	0.23	1.81	1.05	31.81	1.73	21.73
BbF	0.57	0.16	0.08	0.12	1.83	0.28	55.43	1.50	33.34
BkF	0.45	0.13	0.07	0.12	1.78	0.52	0.08	1.50	23.78
BaP	0.54	0.11	0.04	0.12	2.10	0.26	19.87	0.00	21.13
IcdP	0.41	0.10	0.03	0.08	1.94	0.13	10.86	0.15	12.00
DahA	0.11	0.01	0.01	0.01	0.16	0.09	11.28	0.18	20.71
BghiP	0.35	0.12	0.03	0.06	1.59	0.13	35.09	0.38	21.79
P15	**29.98**	**6.37**	**5.17**	**6.77**	**47.27**	**40.89**	**308.52**	**87.60**	**405.40**

Table S3. Standard deviations of emission factors (mg/kg) of individual PAH, total 15 PAHs and BaPeq for different fuels. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC).

	CR	CP	WP	FW	BW	AB	BB	AC	BC
ACY	2.44	0.99	0.51	0.90	6.34	25.21	5.94	10.27	38.61
ACE	4.77	0.08	0.06	0.08	0.16	6.23	6.06	4.04	13.32
FLO	5.70	0.20	0.23	0.24	1.26	13.36	34.50	10.26	28.22
PHE	7.47	1.23	1.80	1.47	5.22	14.12	50.53	12.51	56.48
ANT	1.11	0.14	0.29	0.28	1.14	4.25	15.64	1.26	30.36
FLA	2.92	0.41	0.61	0.61	8.30	6.62	20.29	0.67	37.67
PYR	2.65	0.28	0.42	0.53	7.73	6.19	22.88	0.37	33.97
BaA	0.54	0.10	0.09	0.20	2.73	1.11	23.40	1.78	33.45
CHR	0.54	0.11	0.11	0.18	1.83	2.10	41.75	1.58	29.96
BbF	0.53	0.08	0.06	0.10	1.91	0.41	71.62	2.34	49.79
BkF	0.41	0.06	0.05	0.09	1.86	0.94	0.11	2.35	35.40
BaP	0.57	0.06	0.04	0.11	2.47	0.26	25.60	0.00	28.06

IcdP	0.37	0.06	0.03	0.08	2.27	0.12	13.19	0.17	11.45
DahA	0.12	0.00	0.01	0.01	0.18	0.17	13.44	0.22	31.90
BghiP	0.32	0.07	0.03	0.05	1.79	0.15	44.17	0.43	30.06
P15	**28.24**	**3.20**	**3.85**	**4.51**	**44.92**	**81.08**	**384.10**	**46.67**	**469.10**

S3. Comparison of EF_{PAHs} among various fuels

Table S4. Statistical probability (*p*) values for the comparison of emission factor of total 15 PAHs (**EF_{P15}**). Means, standard deviations, standard errors, and geo-means of EF_{P15} for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC).

	N	Mean ± std	S.E.	Geo-mean	WP	CP	FW	AB	CR	BW	BB	AC	BC
WP	11	5.17±3.85	1	3.99									
CP	12	6.37±3.20	1	5.67	0.342								
FW	66	6.77±4.51	1	5.70	0.218	0.987							
AB	5	40.9±81.1	36	9.13	0.085	0.315	0.254						
CR	25	30.0±28.4	6	19.8	0.000	0.000	0.000	0.077					
BW	10	47.3±44.9	14	34.5	0.000	0.000	0.000	0.007	0.094				
BB	4	309±384	192	69.7	0.000	0.000	0.000	0.001	0.009	0.182			
AC	4	87.6±46.7	23	80.0	0.000	0.000	0.000	0.000	0.004	0.110	0.826		
BC	11	405±469	141	201.0	0.000	0.000	0.000	0.000	0.000	0.000	0.042	0.076	

S4. Comparison of predicted and measured EF_{P15} for biofuel

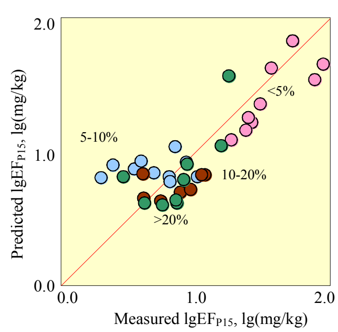

Figure S2. Relationship between the predicted and measured EF_{P15} for biomass fuels. Various fuels are classified into four groups with different moisture contents of < 5 (pink), 5-10 (light blue), 10-20 (brown), and >20% (green).

S5. Dependence of EF_{PAHs} on coal VM content

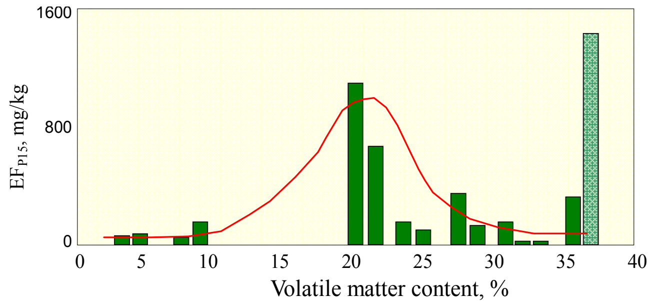

Figure S3. The dependence of EF_{P15} of coal chunks on volatile matter content.

S6. Composition profiles of PAHs in emissions from different fuel combustions

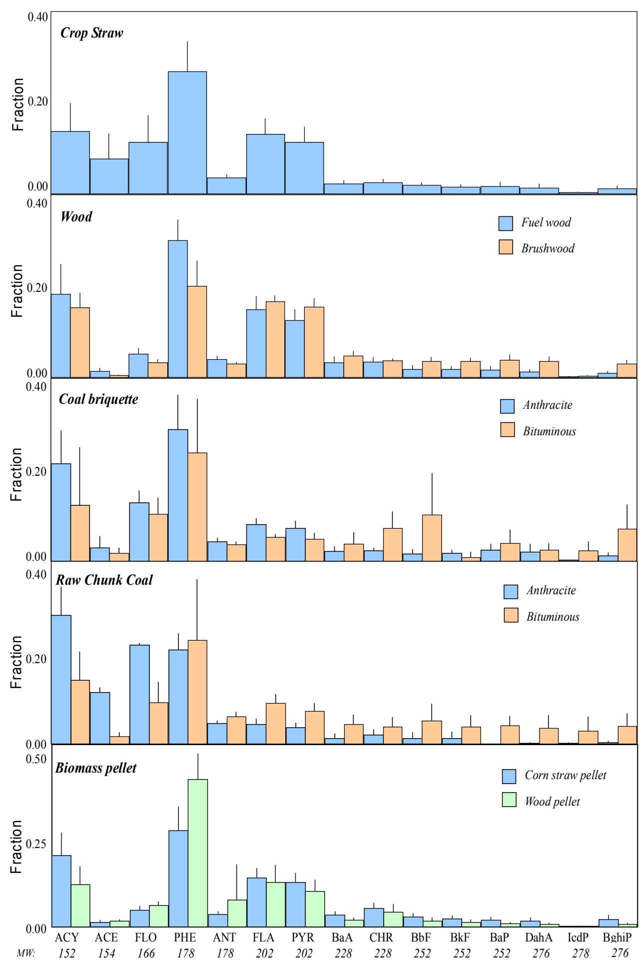

Figure S4 PAH composition profiles from combustion of various fuels in residential stoves. The profiles can be classified into two categories (color-coded as brown and blue/green) by cluster analysis. Data shown are means and standard deviations for each fuel type.

S7. PCA and cluster analysis of composition profile

The principal component analysis extracted four principal factors. A total of four components, explaining over 80% of total variations, are resolved with F1 (8 high molecular weight PAHs from BaA to BghiP, and PHE), F2 (ANT, FLA, and PYR), F3 (ACE and FLO) and F4 (ACY). F1 is mainly associated with high molecular weight PAHs. **Figure S5** shows the loading scores of F1 for different fuel types. BW, BC and BB had relatively higher scores of F1 indicating high contribution of high molecular weight PAHs in emissions from the burning of these three type fuels. The cluster analysis results using the F1 score for various fuels also suggested that these three fuels differed from the other six fuel types (**Figure S5-right panel**).

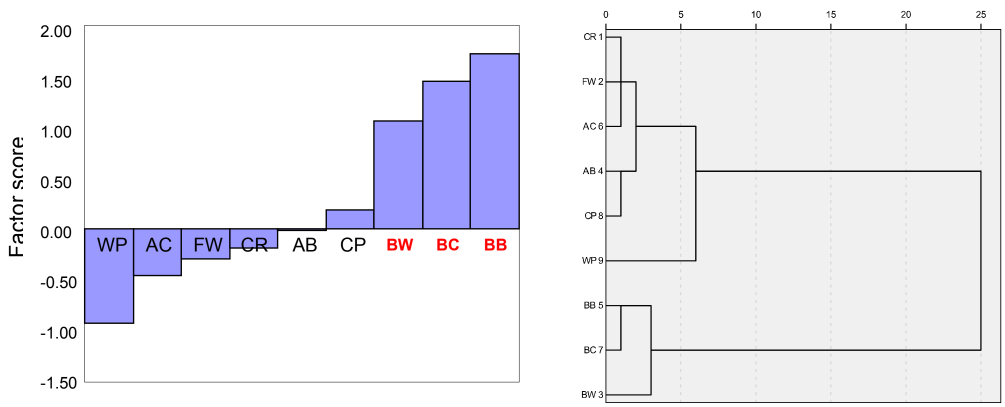

Figure S5. **Loading scores** of **F1** for different fuels (left) and the cluster analysis result based on F1 values of these fuels (right). Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). Brushwood and bituminous coals have high loading scores of F1, while the others have relatively high loadings of F2 and F3.

S8. Comparison of F_{228} among different fuels

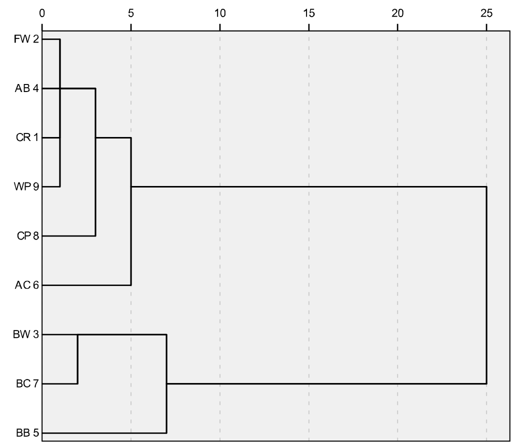

Figure S6. Cluster analysis result of **F_{228}** for different fuels. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC).

Table S5. Statistical probability (*p*) values for the comparison of mass fraction of high molecular weight PAHs (> 228) (**F_{228}**). Means and standard deviations of F_{228} for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC).

	%	N	AC	WP	CR	FW	AB	CP	BW	BC	BB
AC	6.2±5.2	4									
WP	11.3±5.1	11	0.273								
CR	12.6±4.1	25	0.133	0.639							
FW	14.0±5.4	66	0.059	0.304	0.481						
AB	14.1±6.3	5	0.138	0.508	0.702	0.962					
CP	19.1±6.6	12	0.006	0.020	0.022	0.041	0.242				
BW	25.9±5.8	10	0.000	0.000	0.000	0.000	0.008	0.048			
BC	30.8±17.8	11	0.000	0.000	0.000	0.000	0.000	0.001	0.157		
BB	38.0±25.8	4	0.000	0.000	0.000	0.000	0.000	0.000	0.011	0.128	

S9. EFs of BaPeq for different fuel types

Table S6. Statistical results of emission factor of BaP equivalent quantity (**EF_{BaPeq}**) for different fuels. Data shown are means, standard deviations, median, and geo-means.

	Mean ± std	Median	Geo-mean

Crop residue		0.90±0.88	0.70	0.52
Firewood	fuel wood	0.19±0.17	0.13	0.14
	brushwood	3.14±3.60	2.28	1.91
Coal briquette	anthracite	0.57±0.81	0.22	0.30
	bituminous	40.51±50.8	28.00	4.19
Raw coal chunk	anthracite	0.76±0.95	0.37	0.44
	bituminous	51.97±73.2	13.01	12.56
Pellet	corn straw pellet	0.19±0.10	0.17	0.17
	wood pellet	0.09±0.07	0.08	0.07

S10. Comparison of selected isomer ratios among different fuel types

Table S7. Statistical probability (*p*) values for the multiple comparison of ratio of **ANT/(ANT+PHE)**. Means and standard deviations of ANT/(ANT+PHE) for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). The result of ANOVA shows $\boldsymbol{p = 3.2\times10^{-21}}$.

		N	WP	CP	FW	CR	BW	AB	BB	AC	BC
WP	0.10±0.02	11									
CP	0.12±0.02	12	0.150								
FW	0.12±0.01	66	0.129	0.747							
CR	0.12±0.02	25	0.162	0.788	0.976						
BW	0.13±0.02	10	0.013	0.241	0.076	0.112					
AB	0.14±0.06	5	0.028	0.263	0.134	0.160	0.864				
BB	0.16±0.09	4	0.001	0.018	0.005	0.007	0.142	0.247			
AC	0.18±0.04	4	0.000	0.001	0.000	0.000	0.018	0.051	0.445		
BC	0.24±0.08	11	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002	

Table S8. Statistical probability (*p*) values for the multiple comparison of ratio of **FLA/(FLA+PYR)**. Means and standard deviations of FLA/(FLA+PYR) for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). The result of ANOVA shows $\boldsymbol{p = 1.1\times10^{-1}}$.

		N	BW	CP	AB	BB	CR	FW	WP	BC	AC
BW	0.52±0.02	10									
CP	0.53±0.04	12	0.556								
AB	0.53±0.03	5	0.405	0.701							
BB	0.53±0.06	4	0.684	0.984	0.748						

CR	0.54±0.03	25	0.091	0.277	0.716	0.465					
FW	0.54±0.02	66	0.025	0.103	0.504	0.308	0.574				
WP	0.54±0.05	11	0.049	0.142	0.447	0.284	0.522	0.759			
BC	0.55±0.02	11	0.007	0.026	0.174	0.105	0.125	0.193	0.446		
AC	0.56±0.02	4	0.024	0.060	0.186	0.119	0.188	0.261	0.412	0.792	

Table S9. Statistical probability (*p*) values for the multiple comparison of ratio of **BaA/(BaA+CHR)**. Means and standard deviations of BaA/(BaA+CHR) for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). The result of ANOVA shows $\boldsymbol{p = 1.6\times10^{-24}}$.

		N	BB	WP	CP	AC	AB	CR	FW	BC	BQ
BB	0.32±0.07	4									
WP	0.34±0.10	11	0.536								
CP	0.38±0.06	12	0.029	0.031							
AC	0.40±0.12	4	0.031	0.046	0.645						
AB	0.47±0.08	5	0.000	0.000	0.002	0.036					
CR	0.48±0.03	25	0.000	0.000	0.000	0.004	0.764				
FW	0.49±0.04	66	0.000	0.000	0.000	0.000	0.367	0.248			
BC	0.54±0.05	11	0.000	0.000	0.000	0.018	0.002	0.000	0.008		
BW	0.56±0.04	10	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.312	

Table S10. Statistical probability (*p*) values for the multiple comparison of ratio of **IcdP/(IcdP+BghiP)**. Means and standard deviations of IcdP/(IcdP+BghiP) for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). The result of ANOVA shows $\boldsymbol{p = 4.4\times10^{-13}}$.

		N	**BB**	**AC**	**CP**	**WP**	**BC**	**CR**	**AB**	**BW**	**FW**
BB	0.28±0.05	4									
AC	0.29±0.04	4	0.833								
CP	0.46±0.05	12	0.000	0.001							
WP	0.48±0.05	11	0.000	0.000	0.375						
BC	0.52±0.21	11	0.000	0.000	0.042	0.256					
CR	0.53±0.03	25	0.000	0.000	0.003	0.059	0.578				
AB	0.54±0.20	5	0.000	0.000	0.043	0.185	0.668	0.951			
BW	0.54±0.01	10	0.000	0.000	0.008	0.079	0.513	0.821	0.921		
FW	0.55±0.03	66	0.000	0.000	0.000	0.006	0.193	0.340	0.676	0.680	

Table S11. Statistical probability (*p*) values for the multiple comparison of ratio of **BbF/(BbF+BkF)**. Means and standard deviations of BbF/(BbF+BkF) for each fuel are also provided. Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet

(WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). The result of ANOVA shows $\boldsymbol{p = 4.6\times10^{-6}}$.

		N	AB	AC	BW	FW	WP	CP	CR	BC	BB
AB	0.46±0.09	5									
AC	0.49±0.03	3	0.397								
BW	0.50±0.03	10	0.006	0.253							
FW	0.51±0.04	66	0.006	0.253	0.511						
WP	0.55±0.06	12	0.000	0.018	0.007	0.007					
CP	0.55±0.03	11	0.000	0.024	0.013	0.013	0.824				
CR	0.55±0.04	25	0.000	0.014	0.000	0.000	0.906	0.886			
BC	0.55±0.07	11	0.000	0.016	0.006	0.006	0.952	0.776	0.850		
BB	0.57±0.01	2	0.003	0.039	0.090	0.090	0.659	0.572	0.603	0.683	

Table S12. Statistical probability ($\boldsymbol{p}$) values for the multiple comparison of ratio of **BaP/(BaP+BghiP)**. Means and standard deviations of BaP/(BaP+BghiP) for each fuel are also provided. Fuels are classified into 8 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), and bituminous chunk (BC). No data available for anthracite chunk (AC).The result of ANOVA shows $\boldsymbol{p = 3.9\times10^{-16}}$.

		N	BB	BC	CP	BW	CR	WP	FW	AB	AC
BB	0.37±0.03	4									
BC	0.47±0.12	11	0.000								
CP	0.49±0.08	12	0.002	0.002							
BW	0.56±0.02	10	0.000	0.728	0.008						
CR	0.58±0.04	25	0.000	0.616	0.000	0.373					
WP	0.59±0.12	11	0.000	0.616	0.000	0.403	0.929				
FW	0.63±0.04	66	0.000	0.005	0.000	0.002	0.002	0.029			
AB	0.69±0.09	5	0.000	0.001	0.000	0.000	0.001	0.002	0.040		
AC											

Table S13. Statistical probability (p) values for the difference between calculated values and the criteria values for **PAH isomer ratios.** Fuels are classified into 9 types of crop residue (CR), corn pellet (CP), wood pellet (WP), fuel wood (FW), brushwood (BW), anthracite briquette (AB), bituminous briquette (BB), anthracite chunk (AC) and bituminous chunk (BC). The tested criteria values for ANT/(ANT+PHE), FLA/(FLA+PYR), BaA/(BaA+CHR), IcdP/(IcdP+BghiP), and BaP/(BaP+BghiP) are 0.10, 0.50, 0.35, 0.50 and 0.38, respectively. For BbF/(BbF+BkF), in addition to 0.57 which is used for automobile emission, value of 0.48 and 0.79 are used in the statistical analysis for biomass and coal, respectively.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)		BaP/(BaP+BghiP)
	0.10	0.50	0.35	0.50	0.48/0.79	0.57	0.38

CR	0.000	0.000	0.000	0.000	0.000	0.019	0.000
FW	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BW	0.000	0.019	0.000	0.000	0.026	0.000	0.000
CP	0.002	0.043	0.070	0.009	0.000	0.022	0.000
WP	0.868	0.025	0.666	0.270	0.002	0.317	0.000
AB	0.223	0.026	0.033	0.718	0.001	0.044	0.001
BB	0.261	0.404	0.461	0.003	0.028	0.758	0.718
AC	0.045	0.007	0.476	0.012	0.002	0.030	No data
BC	0.000	0.000	0.000	0.783	0.000	0.412	0.000

S11. Correlation among selected isomer ratios for each fuel

Table S14. Correlation coefficients and associated analysis probabilities among different isomer ratios for the anthracite briquette in this study.

	ANT/(ANT+PHE)	**FLA/(FLA+PYR)**	**BaA/(BaA+CHR)**	**IcdP/(IcdP+BghiP)**	**BbF/(BbF+BkF)**	**BaP/(BaP+BghiP)**
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	.100	1.000				
	.873	.				
BaA/(BaA+CHR)	-.300	.100	1.000			
	.624	.873	.			
IcdP/(IcdP+BghiP)	-.300	.100	1.000	1.000		
	.624	.873	.	.		
BbF/(BbF+BkF)	-.300	-.100	.700	.700	1.000	
	.624	.873	.188	.188	.	
BaP/(BaP+BghiP)	-.200	.100	-.200	-.200	.500	1.000
	.747	.873	.747	.747	.391	.

Table S15. Correlation coefficients and associated analysis probabilities among different isomer ratios for the bituminous briquette in the present study.

	ANT/(ANT+PHE)	**FLA/(FLA+PYR)**	**BaA/(BaA+CHR)**	**IcdP/(IcdP+BghiP)**	**BbF/(BbF+BkF)**	**BaP/(BaP+BghiP)**
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	-.800	1.000				
	.200	.				
BaA/(BaA+CHR)	**1.000**	-.800	1.000			
	.	.200	.			

IcdP/(IcdP+BghiP)	-.600	.800	-.600	1.000		
	.400	.200	.400	.		
BbF/(BbF+BkF)	**1.000**	**-1.000**	**1.000**	**1.000**	1.000	
	.	**.**	**.**	**.**	.	
BaP/(BaP+BghiP)	-.800	.400	-.800	.000	-1.000	1.000
	.200	.600	.200	1.000	.	.

Table S16. Correlation coefficients and associated analysis probabilities among different isomer ratios for the anthracite chunk in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)
ANT/(ANT+PHE)	1.000				
	.				
FLA/(FLA+PYR)	**-1.000**	1.000			
	.	.			
BaA/(BaA+CHR)	-.400	.400	1.000		
	.600	.600	.		
IcdP/(IcdP+BghiP)	-.500	.500	.500	1.000	
	.667	.667	.667	.	
BbF/(BbF+BkF)	.000	.000	.000	-.866	1.000
	1.000	1.000	1.000	.333	.

Table S17. Correlation coefficients and associated analysis probabilities among different isomer ratios for the bituminous chunk in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)	BaP/(BaP+BghiP)
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	-.455	1.000				
	.160	.				
BaA/(BaA+CHR)	.055	.591	1.000			
	.873	.056	.			
IcdP/(IcdP+BghiP)	**-.661**	**.806**	.424	1.000		
	.027	**.003**	.194	.		
BbF/(BbF+BkF)	.392	.105	.005	-.196	1.000	
	.233	.759	.989	.563	.	
BaP/(BaP+BghiP)	-.555	.355	.200	**.642**	**-.838**	1.000
	.077	.285	.555	**.033**	**.001**	.

Table S18. Correlation coefficients and associated analysis probabilities among different isomer ratios for the brushwood in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)	BaP/(BaP+BghiP)
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	**-.638**	1.000				
	.047	.				
BaA/(BaA+CHR)	.598	**-.687**	1.000			
	.068	**.028**	.			
IcdP/(IcdP+BghiP)	**.746**	-.360	**.713**	1.000		
	.013	.307	**.021**	.		
BbF/(BbF+BkF)	-.085	.261	.018	.378	1.000	
	.815	.467	.960	.281	.	
BaP/(BaP+BghiP)	.195	.176	.036	.591	**.648**	1.000
	.590	.627	.920	.072	**.043**	.

Table S19. Correlation coefficients and associated analysis probabilities among different isomer ratios for the fuel wood log in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)	BaP/(BaP+BghiP)
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	-.233	1.000				
	.059	.				
BaA/(BaA+CHR)	.569	-.296	1.000			
	.000	.016	.			
IcdP/(IcdP+BghiP)	**.574**	-.130	.482	1.000		
	.000	.299	.000	.		
BbF/(BbF+BkF)	.071	**.292**	-.014	**.264**	1.000	
	.573	**.018**	.912	**.032**	.	
BaP/(BaP+BghiP)	**.376**	.068	.189	**.676**	.199	1.000
	.002	.588	.129	**.000**	.109	.

Table S20. Correlation coefficients and associated analysis probabilities among different isomer ratios for the crop residue in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)	BaP/(BaP+BghiP)
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	**-.406**	1.000				
	.044	.				

BaA/(BaA+CHR)	**.401**	-.065	1.000			
	.047	.759	.			
IcdP/(IcdP+BghiP)	.184	**-.533**	.031	1.000		
	.379	**.006**	.884	.		
BbF/(BbF+BkF)	.204	.296	**.410**	-.271	1.000	
	.328	.150	**.042**	.190	.	
BaP/(BaP+BghiP)	**.474**	**-.450**	.089	.164	-.282	1.000
	.017	**.024**	.671	.433	.172	.

Table S21. Correlation coefficients and associated analysis probabilities among different isomer ratios for the wood pellet in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)	BaP/(BaP+BghiP)
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	.043	1.000				
	.907	.				
BaA/(BaA+CHR)	.371	.164	1.000			
	.291	.631	.			
IcdP/(IcdP+BghiP)	-.201	-.515	.364	1.000		
	.577	.105	.270	.		
BbF/(BbF+BkF)	.000	-.045	-.227	-.333	1.000	
	1.000	.894	.502	.318	.	
BaP/(BaP+BghiP)	**-.742**	.109	.036	.223	.055	1.000
	.014	.750	.915	.509	.873	.

Table S22. Correlation coefficients and associated analysis probabilities among different isomer ratios for the crop straw pellet in the present study.

	ANT/(ANT+PHE)	FLA/(FLA+PYR)	BaA/(BaA+CHR)	IcdP/(IcdP+BghiP)	BbF/(BbF+BkF)	BaP/(BaP+BghiP)
ANT/(ANT+PHE)	1.000					
	.					
FLA/(FLA+PYR)	-.175	1.000				
	.586	.				
BaA/(BaA+CHR)	-.287	-.277	1.000			
	.366	.384	.			
IcdP/(IcdP+BghiP)	**-.678**	.413	.084	1.000		
	.015	.182	.795	.		
BbF/(BbF+BkF)	**.571**	-.342	-.532	-.186	1.000	
	.053	.276	.075	.564	.	

BaP/(BaP+BghiP)	.077	.158	-.056	**.594**	.462	1.000
	.812	.625	.863	**.042**	.130	.

S12. PAH phase distribution in emissions for different fuels

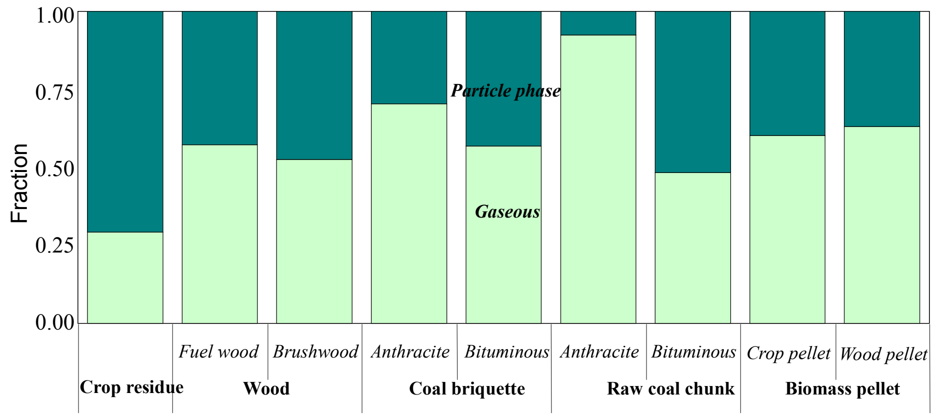

Figure S7. Phase distribution of **total PAHs** for different fuels. Fuels are classified into nine types of crop residue, corn pellet, wood pellet, fuel wood, brushwood, anthracite briquette, bituminous briquette, anthracite chunk and bituminous chunk.

S13. Size distribution of particle-phase PAHs in emissions for different fuels

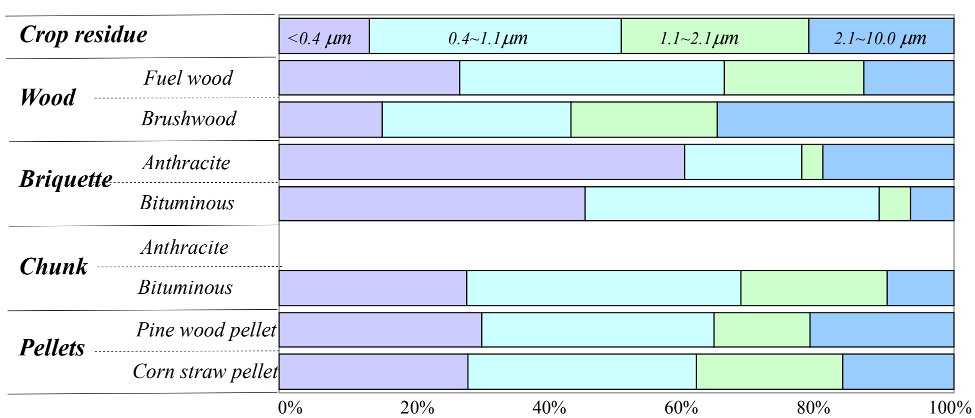

Figure S8. Size distribution of **particle-bound PAHs** for different fuels. Fuels are classified into 9 types of crop residue, corn pellet, wood pellet, fuel wood, brushwood, anthracite briquette, bituminous briquette, and bituminous chunk. Data for anthracite chunk is not available. Particulate matter is segregated into 4 fractions of particle with diameter less than 0.4, 0.4-1.1, 1.1-2.1 and 2.1-10 μm, respectively.

References

Shen, G. F.; Yang, Y. F.; Wang, W.; Tao, S.; Zhu, C.; Min, Y.; Xue, M.; Ding, J.; Wang, B.; Wang, R.; Shen, H.; Li, W.; Wang, X.; Russell, A. G. Emission factors of particulate matter and elemental carbon for crop residues and coals burned in typical household stoves in China. *Environ. Sci. Technol*. **2010**, *44,* 7157-7162.

Shen, G.; Tao. S.; Wei. S.; Zhang, Y.; Wang, R.; Wang, B.; Li, W.; Shen, H.; Huang, Y.; Yang, Y.; Wang, W.; Wei, W.; Wang, X.; Liu, W; Wang, X.; Simonich, S. Reductions in emissions of carbonaceous particulate matter and polycyclic aromatic hydrocarbons from combustion of biomass pellets in comparison with raw fuel burning. *Environ. Sci. Technol.* **2012,** *46,* 6409-6416.