Supporting Information for "Field Measurement on the Emissions of PM, EC, OC, Parent, Nitro- and Oxy- Polycyclic Aromatic Hydrocarbons from Residential Briquette, Coal Cake, and Wood Combustions in Rural Shanxi, China"

Guofeng SHEN[1,2], Shu TAO[1*], Siye WEI[1], Yuanchen CHEN[1],Yanyan ZHANG[1], Huizhong SHEN[1], Ye HUANG[1], Dan ZHU[1], Chenyi YUAN[1], Haochen WANG[3], Yafei Wang[3], Lijun PEI[3], YiLan LIAO[3], Yonghong DUAN[4], Bin WANG[1], Rong WANG[1], Yan Lv[1], Wei LI[1], Xilong WANG[1]

1. *Laboratory of Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing, 100871, China*
2. *Institute of Atmospheric Sciences, Jiangsu Academy of Environmental Sciences, Nanjing, 210036, China*
3. *Institute of Population Research, Peking University, Beijing 100871, China*
4. *College of Resources and Environment, Shanxi Agricultural University, Shanxi, 030800, China*

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

There are 5 tables and 2 figures

Figure S1. Pictures of the fuel and stove tested in this study. Three different fuel/stove combinations were inventigated: wood burned in a simple metal stove, honmeycomb briquette burned in an improved metallic stove with a chimney, and coal cake burned in a brick stove with a outdoor flue pipe (from left to the right).

Table S1. Fuel moisture (%, wet basis) and proximate analysis (%, dry basis) for briquette, coal cake and wood investigated in this study.

	Briquette	Coal cake	Wood
Moisture	1.5	2.1	5.4
Volatile matter	11	6.1	83
Ash	47	87	1.0
Fixed carbon	42	6.8	16

The temperature protocol used in EC/OC analysis is listed in **Table S2**. The protocol used here was a modified one similar to the National Institute of Occupational Safety and Health (NIOSH) 5040, as recommended by the manufacturer. Briefly, it was increased to 600 (90 s), 840 ^{o}C (90 s) in a pure helium atmosphere for OC detection, and then 550 (35 s), 650 (45 s) and 870 (90 s) ^{o}C in an oxygen/helium atmosphere for EC detection.

Table S2. EC/OC analysis temperature protocol (rtquartz.par) in this study using a Sunset analyzer (RT-4)

Mode	Time, s	Temperature, ^{o}C	Power constant	Time constant, s	Blower mode
Helium	10	1	0.001	95	0
Helium	90	600	20	40	0
Helium	90	840	32	22	0
Helium	35	0	0.001	95	16
Helium	1	550	20	50	0
Oxygen	35	550	18	50	0
Oxygen	45	650	22	35	0
Oxygen	90	870	32	22	0

CalibrationOx	110	0	0.001	95	16
OFFLINE	1	0	0.001	95	16

Table S3. Instrumental detection limit (defined as 3 times signal-to-noise ratio), method detection limit (defined as 3.14 times the standard deviation of 7 replicated analyses of spiked samples) and recoveries of individual parent PAHs, oxy- and nitro-PAHs.

	IDL, ng	MDL, ng/mL		Recovery	
		gaseous	particle phase	gaseous	particle phase
Parent-PAHs					
naphthalene (NAP)	0.40	0.99	0.32	0.70	0.74
acenaphthylene (ACY)	0.13	0.63	0.24	0.83	0.68
acenaphthene (ACE)	0.43	0.49	0.31	0.73	0.71
fluorene (FLO)	0.76	0.64	0.25	0.95	0.83
phenanthrene (PHE)	0.78	0.53	1.18	0.89	0.73
anthracene (ANT)	0.48	0.84	0.50	1.01	0.77
fluoranthene (FLA)	0.31	1.26	0.91	1.14	0.78
pyrene (PYR)	0.38	1.26	0.95	1.21	0.80
retene (RET)	0.31	0.74	0.40	1.04	0.85
benzo[c]phenanthrene (BcP)	0.35	0.88	0.54	0.89	0.90
cyclopenta[c,d]pyrene (CPP)	0.40	0.90	0.50	0.92	0.90
benzo(a)anthracene (BaA)	0.35	0.97	0.67	0.85	0.85
chrysene (CHR)	0.41	1.06	0.77	0.90	0.98
benzo(b)fluoranthene (BbF)	0.30	0.91	1.20	0.80	1.21
benzo(k)fluoranthene (BkF)	0.48	1.71	1.23	1.08	0.99
benzo(a)pyrene (BaP)	0.47	1.43	1.03	1.01	0.80
benzo(e)pyrene (BeP)	0.47	1.43	1.02	0.95	0.80
perylene (PER)	0.23	0.68	1.02	0.94	0.79
indeno(l,2,3-cd)pyrene (IcdP)	0.31	0.82	1.05	0.72	1.20
dibenz(a,h)anthracene (DahA)	0.93	1.28	0.98	0.83	1.03
benzo(g,h,i)perylene (BghiP)	0.93	1.32	1.42	0.90	0.97
dibenzo[a,c]pyrene (DacP)	0.90	1.10	1.30	0.80	0.96
dibenzo[a,l]pyrene (DalP)	0.84	1.01	0.92	0.75	0.99
dibenzo[a,e]flluoranthene (DaeF)	0.85	1.00	1.02	0.78	1.05
Coronene(COR)	0.78	0.99	1.04	0.98	0.98
dibenzo[a,e]pyrene (DaeP)	0.89	1.33	1.25	0.86	1.35
dibenzo[a,i]pyrene (DaiP)	0.92	1.03	1.10	0.88	1.00
dibenzo[a,h]pyrene (DahP)	0.90	1.05	1.08	0.90	0.98
Oxy-PAHs					
9-fluorenone (9FO)	0.06	0.43	0.08	0.72	0.53
anthracene-9,10-dione (ATQ)	0.24	0.84	0.30	0.96	0.81
benzanthrone (BZO)	0.13	0.54	0.44	0.84	0.87
benzo[a]anthracene-dione(BaAQ)	0.12	0.59	0.24	0.80	0.71
Nitro-PAHs					
1-nitro-naphthalene (1N-NAP)	0.12	0.44	0.67	0.72	0.83

2-nitro-naphthalene (2N-NAP)	0.24	0.57	0.71	0.83	0.84
5-nitro-acenaohthene (5N-ACE)	0.12	0.58	0.75	0.77	0.88
2-nitro-fluorene (2N-FLO)	0.24	0.64	0.75	0.83	0.90
9-nitro-anthracene (9N-ANT)	0.12	0.56	0.64	0.68	0.76
9-nitro-phenanthrene (9N-PHE)	0.12	0.56	0.73	0.71	0.86
3-nitro-phenanthrene (3N-PHE)	0.13	0.71	0.74	0.79	0.90
3-nitro-fluoranthene (3N-FLA)	0.24	0.75	0.74	0.82	0.87
1-nitro-pyrene (1N-PYR)	0.12	0.67	0.82	0.78	0.83

Table S4. EFs (mg/kg) of individual parent PAHs, oxy- and nitro-PAHs from residential briquette, coal cake, and wood combustions. Data shown are the range from duplicate measurements.

	Briquette	Coal Cake	Wood
Parent-PAHs			
naphthalene (NAP)	2.2-3.2	18-87	9.3-172
acenaphthylene (ACY)	0.59-0.82	32-42	15-28
acenaphthene (ACE)	0.052-0.086	1.7-2.1	1.3-2.0
fluorene (FLO)	0.29-0.42	9.5-9.6	4.1-4.9
phenanthrene (PHE)	3.0-3.5	26-29	16-22
anthracene (ANT)	0.33-0.38	8.1-9.1	3.5-4.6
fluoranthene (FLA)	2.2-2.3	11-14	14-26
pyrene (PYR)	1.6-1.7	8.6-11	13-23
retene (RET)	0.058-0.11	0.70-0.85	0.88-8.1
benzo[c]phenanthrene (BcP)	0.14-0.16	0.99-1.5	1.0-1.9
cyclopenta[c,d]pyrene (CPP)	0.022-0.11	7.8-12	12-22
benzo(a)anthracene (BaA)	0.58-0.74	4.3-6.6	4.5-8.1
chrysene (CHR)	1.2-1.3	2.8-4.2	3.6-6.6
benzo(b)fluoranthene (BbF)	0.31-0.38	2.0-2.9	2.7-4.2
benzo(k)fluoranthene (BkF)	0.24-0.30	1.8-2.5	3.0-4.5
benzo(a)pyrene (BaP)	0.20-0.22	0.95-1.3	1.7-2.6
benzo(e)pyrene (BeP)	0.068-0.077	2.4-3.4	3.8-5.9
perylene (PER)	0.0094-0.0096	0.43-0.55	0.62-0.96
indeno(l,2,3-cd)pyrene (IcdP)	0.034-0.035	1.8-2.7	2.7-3.9
dibenz(a,h)anthracene (DahA)	0.018-0.019	0.19-0.32	0.33-0.39
benzo(g,h,i)perylene (BghiP)	0.033-0.034	0.86-1.3	1.9-2.7
dibenzo[a,c]pyrene (DacP)	0.0014-0.0018	0.67-0.67	0.78-1.5
dibenzo[a,l]pyrene (DalP)	0.0050-0.0067	0.41-0.58	0.36-0.55
dibenzo[a,e]flluoranthene (DaeF)	0.0022-0.0039	0.31-0.39	0.33-0.37
Coronene(COR)	0.0055-0.0066	0.75-1.2	2.2-2.6
dibenzo[a,e]pyrene (DaeP)	0.0084-0.012	0.23-0.39	0.16-0.39
dibenzo[a,i]pyrene (DaiP)	0.0014	0.27-0.47	0.22-0.45
dibenzo[a,h]pyrene (DahP)	n.d.*	0.048-0.067	0.053-0.097
Oxy-PAHs			
9-fluorenone (9FO)	1.1-1.8	3.4-4.1	2.9-4.1

anthracene-9,10-dione (ATQ)	0.50-0.68	0.69-1.5	1.5-2.0
benzanthrone (BZO)	0.077-0.16	0.65-3.8	3.44-3.89
benzo[a]anthracene-dione(BaAQ)	0.014-0.018	0.018-0.062	0.096-0.15
Nitro-PAHs			
1-nitro-naphthalene (1N-NAP)	0.00038-0.00049	0.0037-0.0041	0.0098-0.011
2-nitro-naphthalene (2N-NAP)	0.00071-0.0012	0.0043-0.0045	0.010-0.011
5-nitro-acenaohthene (5N-ACE)	0.00032-0.00062	0.0011-0.0033	0.0054-0.0070
2-nitro-fluorene (2N-FLO)	0.00063-0.00067	0.0012-0.0033	0.0047-0.0068
9-nitro-anthracene (9N-ANT)	0.47-0.59	0.13-2.1	0.04-0.33
9-nitro-phenanthrene (9N-PHE)	0.16-0.24	0.016-0.35	0.042-0.18
3-nitro-phenanthrene (3N-PHE)	0.00022-0.00025	0.00030-0.00088	0.0015-0.0020
3-nitro-fluoranthene (3N-FLA)	0.00073-0.00075	0.00057-0.0017	0.0052-0.010
1-nitro-pyrene (1N-PYR)	0.0011-0.0017	0.0030-0.0053	0.0059

*: n.d. not detected.

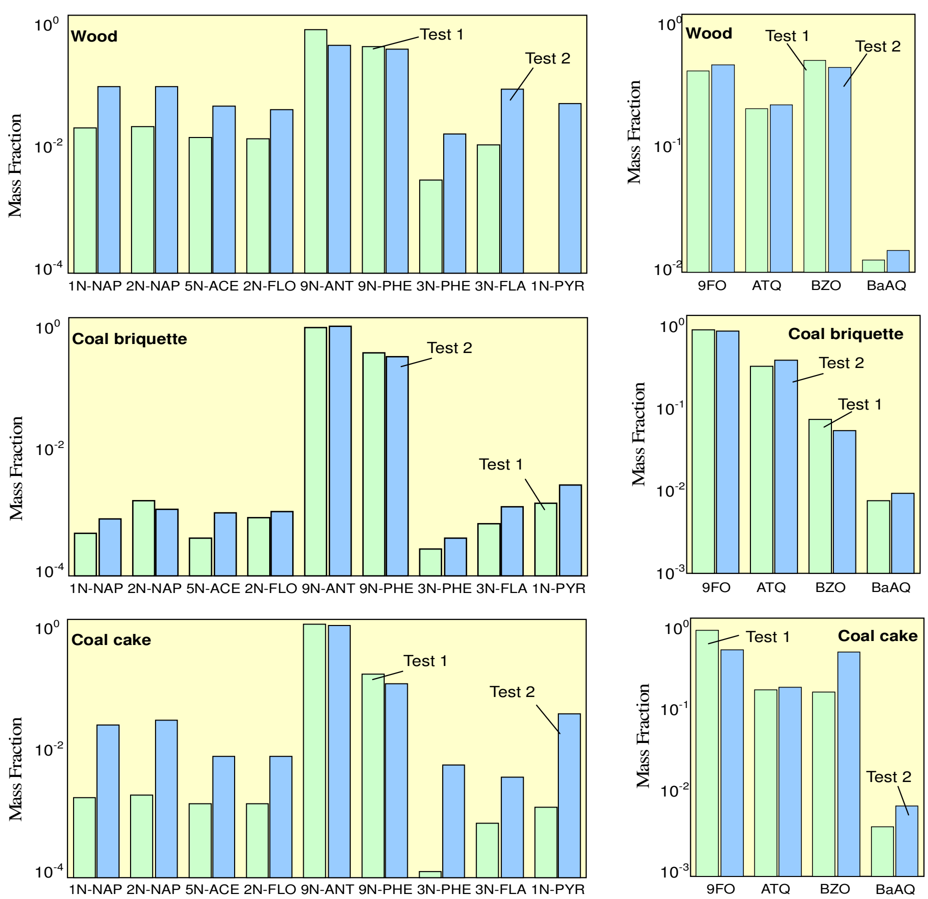

Figure S2. Normalized mass percentages of 4 oxygenated and 9 nitro PAHs (gaseous and particle-phase combined) for the wood, coal cake and coal briquette, respectively.

Table S5. Comparison of EFs of PM, OC, EC, parent PAHs and their derivatives measured from residential wood and coal combustion in rural China.

Fuel	Stove	PM	OC	EC	pPAHs	oPAHs	nPAHs	reference
		g/kg	g/kg	g/kg	mg/kg	mg/kg	mg/kg	
Field measurement								
Wood	A movable metal stove without chimney	8.1-8.5	2.2-3.6	0.91-1.6	182-297 [1]	7.8-10	0.14-0.55	This study
	An improved stove with two pot, no grate	2.95-4.58[2]	0.41-1.43	1.03-3.26				Li *et al.*, 2009
	An improved stove with one pot, bottom grate	2.67±0.42[2]	1.07±0.16	1.14±0.24				Li *et al.*, 2009
	Stove/kang, one pot, no grate with a flue [3]	3.04±0.85[2]	0.98±0.32	1.32±0.39				Li *et al.*, 2009
Briquette	An improved stove with a chimney	0.54-0.64	0.13-0.14	0.0040-0.0041	14-16[4]	1.7-2.6	0.64-0.83	This study
Coal cake	A brick stove with an outdoor flue	3.2-8.5	0.38-0.58	0.022-0.052	168-223[4]	4.7-9.5	0.16-2.4	This study
Combustion under laboratory conditions								
Wood	Brick stove with a flue	1.54±0.20						Zhang *et al.*, 2000

	Improved brick stove with a flue	4.10±1.5						Zhang *et al.*, 2000
	Metal stove without flue	2.67±1.20						Zhang *et al.*, 2000
	A typical brick stove/kang with one pot and an outdoor flue	1.6±0.32	0.60±0.35	0.94±0.40	6.4±2.8[4]	0.37±0.27	0.0051±0.0029	Shen *et al.*, 2012
Briquette	Metal stove without flue	0.27±0.073						Zhang *et al.*, 2000
	Metal stove with a flue	0.22±0.11						Zhang *et al.*, 2000
	Improved metal stove without flue	0.48±0.57						Zhang *et al.*, 2000
	Metal stove with a flue[5]	0.62±0.73						Zhang *et al.*, 2000
	A metallic outer cover and thermal-insulated ceramic liner[6]	1.33-19.6	0.017-13.8	0.004-0.675	69.0-160[7]			Chen *et al.*, 2005
	A traditional portable stove	2.78-26.4	1.91-13.0	0.044-0.64				Zhi *et al.*, 2008, 2009
	An improved stove with an upper lid and a	2.54-9.85	1.02-6.17	0.011-0.16				Zhi *et al.*, 2008, 2009

	flue							
	Stoves with single or double flues				186-812[8]			Liu *et al.,* 2009
	A metallic outer cover and thermal-insulated ceramic liner				0.049[7]			Chen *et al.,* 2004
	A movable metallic stove without flue	0.17±0.010	0.021±0.0020	0.0042±0.0021	14±3.1[9]	0.29±0.016		Shen *et al.,* 2010a, 2010b
	A Stove with a flue, a cylinder 24 cm in diameter and 50 cm in height	0.614±0.0248			0.011±0.0093 (only BaP) [10]			Ge *et al.*, 2004
Coal cake	A Stove with a flue, a cylinder 24 cm in diameter and 50 cm in height	0.995±0.0609			0.0021±0.0022 (only BaP)			Ge *et al.*, 2004

[1] EFs of a total of 28 PAHs analyzed by GC-MS. Results are gas- and particle-phase combined.

[2] $PM_{2.5}$ measured in the study

[3] The fuel was branch

[4] Analysis methods were the same as those for the wood, and the results are gas- and particle-phase combined.

[5] Honeycomb coal from a special coal mine in Shanxi

[6] A variety of coals (1 anthracite, 1 sub-bituminous and 3 bituminous coals) were burned.
[7] 14 parent PAHs in both gaseous and particle phases.
[8] 15 PAHs (except for NAP) were analyzed by GC-MS. Results are total PAHs in both gaseous and particulate phases.
[9] 16 PAHs in both gaseous and particle phases. Analytical methods were the same as those in this study.
[10] BaP was analyzed by HPLC. Results are total BaP in both gaseous and particle phases.

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