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Increase in polycyclic aromatic hydrocarbon (PAH) emissions due to briquetting: A challenge to the coal briquetting policy



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ABSTRACT

Both China and UNEP recommend replacing raw coal chunks with coal briquettes in household sector as clean coal technology (CCT), which has been confirmed by the decreased emissions of particulate matter and black carbon. However, the clean effect has never been systematically checked by other pollutants like polycyclic aromatic hydrocarbons (PAHs). In this study, 5 coals with different geological maturities were processed as both chunks and briquettes and burned in 3 typical coal stoves for the measurement of emission factors (EFs) of particle-bound PAHs. It was found that the EFs of 16 parent PAHs, 26 nitrated PAHs, 6 oxygenated PAHs, and 8 alkylated PAHs for coal briquettes were 6.90 ± 7.89 , 0.04 ± 0.03 , 0.65 ± 0.40 , and 72.78 ± 18.23 mg/kg, respectively, which were approximately 3.1, 3.7, 1.9, and 171 times those for coal chunks, respectively. Such significant increases in PAH emissions increased human health risk and challenged the policy of CCT.

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1. Introduction

As industrialization and urbanization continues in China, energy consumption grows concomitantly. Regional air quality problems have become increasingly prominent, typified by more frequent occurrences of sustained heavy fog-haze events, which increasingly harms human health and even affects social harmony and stability (Chen et al., 2013; MEP, 2013). To reverse this trend of pollution, in September 2013 China's central government issued the *Action Plan of Air Pollution Prevention and Control* (referred to as the *Action Plan* hereafter). According to the *Action Plan*, by 2017 the urban concentration of inhalable particulate matter (PM₁₀) shall decrease by 10% compared with 2012 and the annual compliant days shall increase year by year (http://www.gov.cn/zwqk/2013-09/12/content_2486773.htm).

The *Action Plan* regards the control of coal smoke emissions as an effective action to reduce ambient and indoor air pollution because of the dominant role of coal in China's energy structure. In 2012, coal consumption reached 3526 million tons (Mt), accounting for approximately 70% of China's total primary energy (NBSC, 2013). Moreover, burning raw coal in residential cooking/heating stoves has the potential to release pollutants that consist of up to 10% of the fuel mass due to poor combustion conditions and control facilities (Zhang and Smith, 2007). Therefore, more effective measures ought to be taken to counteract the adverse effects of coal consumption on both outdoor and indoor air quality.

Since the 9th Five-Year Plan period (1996–2000), a strategy for the development and deployment of clean coal technology (CCT) for the purpose of efficient and clean combustion has been formulated in China (Cheng, 1998). One of the CCTs is the transformation of raw coal chunks into honeycomb coal briquettes (i.e., briquetting). According to the *9th Five-Year Plan and 2010 Development Outline for the National Clean Coal Technology* (www.docin.com/p-441441349.html, in Chinese), coal briquettes have twice the

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combustion efficiency and result in a 20–30% saving of coal consumption, a 40–60% decrease in smoke and SO₂ emissions, and an 80% decline in CO release compared with raw coal chunks. Our previous study presented corroborative evidence that PM and black carbon (BC) emissions from briquette combustion in a traditional stove could be reduced by $34 \pm 15\%$ and $90 \pm 6\%$, respectively, compared to raw coal chunk combustion in the same stove (Zhi et al., 2009). In the *Action Plan*, the vigorous promotion of coal briquettes is particularly emphasized as a CCT for the reduction of air pollutant emissions from coal combustion in the residential sector.

The benefits of coal briquetting have been expanded from clean air purposes to climate mitigation. Although the control of BC emissions has long been considered as useful for climate mitigation, the unavoidable co-control of other pollutants such as organic carbon (OC) and inorganic aerosols (which mostly contribute to climate cooling) often causes uncertainty as to whether the net effect would be climate cooling or warming (Bond et al., 2013). Fortunately, converting raw coal chunks to coal briquettes has been considered as net-cooling activity and effective against both anthropogenic global warming and air pollution. In 2011, the UNEP/WMO published the *Integrated Assessment of Black Carbon and Tropospheric Ozone*, recommending “replacing (raw) coal with coal briquettes for cooking and heating stoves” as the top priority in residential sector for its co-benefits to public health and climate mitigation (UNEP and WMO, 2011). This recommendation was further supported by some subsequent assessment reports associated with short-lived climate pollutants (SLCPs) (e.g., UNEP, 2011; World Bank, 2013), which boosted the confidence regarding the co-benefits for health and climate from the replacement of raw coal chunks with briquettes.

However, recently reported measurements on biomass pellets (biomass briquettes) showed contradictory results. Emissions of PCDD/Fs and PCBs were observed to be unexpectedly high during the combustion of wood (intermittent) and straw (continuous) pellets (Hedman et al., 2006), and the emissions of some toxic organic compounds, such as benzene, naphthalene, and furan were found to be higher in the after-flame phase of pellet burning than in the combustion of raw oat straw (Perzon, 2010). More noteworthy, Shen et al. (2012) found that the briquetting of biomass on one hand led to lower emission factors (EFs) for CO, OC, BC, and PM, but on the other hand likely led to elevated EFs for polycyclic aromatic hydrocarbons (PAHs). PAHs are among the most toxic organic pollutants due to their carcinogenic, mutagenic, and teratogenic properties (Luch, 2005; Mumford et al., 1987; Zhang and Smith, 2007), and exposure to high levels of PAHs is believed to be related to increased cancer risk and adverse birth outcomes (Boström et al., 2002; Perera et al., 2009; Zhang et al., 2009). The possible increase in PAHs and other organic pollutants from the burning of biomass pellets mentioned above implies the likelihood that coal briquettes may also have higher rather than the presumed lower PAH emissions than raw coal chunks. If this is true, including coal briquetting in CCTs and climate-health co-benefit efforts is somewhat misleading or less convincing, and the current policy of replacing coal chunks with coal briquettes for cleaner air, better public health and climate benefits must be reconsidered in a more comprehensive way.

The present study intends to (i) clarify whether coal briquetting increases PAH emissions under household burning conditions, (ii) serve as a preliminary probe into factors accounting for the putative PAH increase, and (iii) identify future research need and policy considerations that could help to reduce PAHs and other emissions such as BC when raw coal chunks are phased out and briquette coal is deployed.

2. Materials and methods

2.1. Coal/stove combinations and sample collection

All materials and procedure used in the experiment of household coal combustion were described in detail previously (Chen et al., 2015); only a brief description will be given here, including five coals, 3 stoves, a large-sized dilution sampling system, and the sample collection procedure.

The five coals covered a wide range of geological maturity, including 1 high volatile bituminous (HVB) coal (YL), 2 medium volatile bituminous (MVB) coals (CX and ZY), 1 low volatile bituminous (LVB) coal (CZ), and 1 semi-anthracite (SA) coal (AY). The vitrinite reflectances (Ro) were 0.72%, 1.00%, 1.50%, 1.90%, and 2.47%, respectively. Each coal was processed into both raw coal chunks and honeycomb briquettes for testing the briquetting effect on PAH EFs. The basic information of the 5 coals is presented in Table S1 in the Supporting Information (SI).

The three stoves selected in this study were designated GC, SC and XF, respectively. The GC stove is a traditional and simple stove extensively used in rural households. The SC and XF stoves are both well-suited for briquette burning, only that the XF stove has a helical inner wall, on which a ring of half-way air-admission holes were arranged to facilitate the supply of air during coal burning; this decided the higher thermal efficiency than SC stove. In the present experiment, GC stove was used only for burning coal chunks, XF stove only for coal briquettes, while SC stove for both chunks and briquettes due to its medium thermal efficiency among them. Thus there were a total of 20 coal/stove combinations in the experiment.

The large-sized stationary full-flow dilution tunnel and fractional sampling system was composed of several parts: a hood and pipe that ducted the smoke from the stove chimney into the dilution tunnel, a set of high-efficiency filters that provided particle-free air for smoke dilution, a seamless stainless-metal tunnel with a mixing ring inside to keep the smoke diluted evenly, and a venturi pump assembly at the end of the tunnel to accurately draw the smoke through the tunnel at a preset flow rate.

A fractional stream of dilution smoke was drawn to quartz-filter samplers equipped with PM_{2.5} cyclones near the end of the tunnel (Fig. S1 in the Supporting Information). The fractional ratio of sampled to total emissions (about 5%) and the coal weight actually burned (exclusive of ash content in chunks and clay added in briquettes) were recorded to facilitate the calculation of EFs of pollutants from coal burning. The calculation of EFs on the basis of fuel weight burned is described in more detail in the Supporting Information.

2.2. PAH analysis and quality control

Fifty-six individual PAHs were measured in each smoke sample, including 16 parent PAHs (pPAHs), 26 nitrated PAHs (nPAHs), 6 oxygenated PAHs (oPAHs), and 8 alkylated PAHs (aPAHs) (Table S2 in SI). The analytical procedures for these PAHs were previously described (Bi et al., 2003; Huang et al., 2014). Briefly, quartz-filter samples of coal smoke were Soxhlet-extracted for 24 h with dichloromethane (DCM) after the surrogate standard mixture was spiked with naphthalene-D8, acenaphthene-D10, phenanthrene-D10, chrysene-D12, perylene-D12, 5-nitroacenaphthene-D9, 9-nitroanthracene-D9, 3-nitrofluoranthene-D9, and 6-nitrochrysene-D11. The extracts were concentrated and fractionated using silica-alumina columns. The fraction containing pPAHs, oPAHs, nPAHs, and aPAHs was collected, reduced almost to dryness and then dissolved in *n*-hexane. Prior to instrumental analysis, known quantities of internal standards, including hexamethyl benzene (for

pPAHs and aPAHs) and 2-nitrofluorene (for oPAHs and nPAHs) were added to the samples. The pretreatment of raw coal samples involved pulverizing coal chunk to powder, passing 100 meshes, weighing 5–10 g of coal powder for Soxhlet-extraction by DCM, and then following the PAH procedure for coal smoke samples described above.

All samples were analyzed using a gas chromatography/mass spectrometer (Agilent 7890A GC/5975C MSD). The mass spectrometer was operated in electron impact ion (EI) mode and selected ion monitoring (SIM) for pPAHs and aPAHs, and in electron capture negative ion (ECNI) mode and SIM for oPAHs and nPAHs. Note that only pPAHs and aPAHs were quantitatively analyzed for raw coal samples.

Quality assurance and quality control (QA/QC) were performed during coal smoke sampling and PAH analysis, including pre-baking of quartz filters at 550 °C before sampling, separate collection of field blank samples and 20% parallel smoke samples, and analysis of more than 10% of parallel samples of coal smoke and coal powder. The recoveries for deuterated PAHs ranged from 75% to 110%, and the PAH concentrations were corrected by the recovery efficiency of the deuterated PAHs.

3. Results and discussion

3.1. Measured increase in PAH emissions due to briquetting

The presentation of each PAH EF in each sample will be made in another paper and the present study mainly emphasizes the general impact of burning briquettes or chunks on PAH emissions. Therefore, we only calculated grouped PAH EFs (i.e., pPAHs, nPAHs, oPAHs, and aPAHs groups) for each of the 20 coal/stove combinations (Table 1). As shown in Table 1, the mean EF values of pPAHs, nPAHs, oPAHs, and aPAHs of five coals were 2.270 ± 1.607 , 0.014 ± 0.012 , 0.418 ± 0.272 , and 0.971 ± 0.627 mg/kg (average \pm standard deviation), respectively, for the burning style of raw chunk, while 6.898 ± 7.893 , 0.043 ± 0.026 , 0.648 ± 0.401 , and 72.777 ± 18.227 mg/kg, respectively, for that of honeycomb briquette. These values generally fell in the ranges of previously

Table 2
Briquette/chunk ratios of OC and grouped PAH emissions.

Coal ID	OC	pPAHs	nPAHs	oPAHs	aPAHs
Briquette/Chunk ratio (in SC stove)					
Coal ID	OC	pPAHs	nPAHs	oPAHs	aPAHs
YL	3.25	8.02	na ^a	na ^a	54.94
CX	2.67	1.33	1.63	1.31	76.39
ZY	2.31	1.33	6.18	1.95	51.94
CZ	0.76	3.18	17.7	2.47	116.57
AY	3.50	5.58	4.43	4.17	609.87
Average	2.50	3.89	7.50	2.47	181.94
SD ^b	1.08	2.90	7.09	1.23	240.60
Briquette/Chunk ratio (in average)					
YL	2.96	6.58	3.63	2.02	56.99
CX	1.80	1.80	2.55	1.39	62.55
ZY	1.34	1.23	4.12	1.67	42.91
CZ	0.83	2.40	3.90	1.45	135.93
AY	2.50	3.33	4.38	2.94	558.19
Avg	1.88	3.07	3.71	1.90	171.31
SD ^b	0.86	2.11	0.71	0.64	219.27

^a Data not available due to failed PAH measurement.

^b Standard deviation.

reported results though were somewhat lower than them (Chen et al., 2005; Huang et al., 2014; Shen et al., 2010, 2013). For example, Huang et al. (2014) reported the EFs of 9.82–215, 0.142–1.88, and 4.47–20.8 mg/kg for 15 pPAHs, 26 nPAHs, and 6 oPAHs, respectively, in total particulate and gaseous phases for a few honeycomb coal briquettes. The higher efficiency stoves used and the less PAH phase (only particle phase, excluding gaseous phase) measured in the present experiment may account for the relatively low emission factors of PAHs. We notice that literature data on grouped or individual PAH EFs from household coal combustion differ in a very wide range, e.g., one or two orders of magnitude and this is usually attributed to the condition-dependent EFs (e.g. PAHs, BC, etc.) for residential coal burning. For example, differences in conditions of stoves, coals, and/or ventilation between experiments could make the differences in EFs

Table 1
Emission factors of grouped PAHs (mg/kg) and OC (g/kg) for household combustion of coal chunks and honeycomb briquettes.

Coal ID	Rank	Stove	Burning style	OC	Σ pPAHs	Σ nPAHs	Σ oPAHs	Σ aPAHs
YL	HVB	GC	Chunk	0.73	3.462	0.005	0.334	1.162
CX	MVB			1.02	3.037	0.011	0.521	1.779
ZY	MVB			1.76	5.176	0.014	0.763	1.767
CZ	LVB			0.28	1.361	0.017	0.341	0.584
AY	SA			0.04	0.245	0.001	0.100	0.144
YL	HVB	SC	Chunk	0.61	2.975	0.027	0.882	1.139
CX	MVB			0.94	2.859	0.040	0.596	1.422
ZY	MVB			0.83	2.693	0.011	0.387	1.213
CZ	LVB			0.25	0.765	0.004	0.202	0.402
AY	SA			0.02	0.129	0.007	0.059	0.097
		Average for chunk		0.648	2.270	0.014	0.418	0.971
		SD ^b		0.533	1.607	0.012	0.272	0.627
YL	HVB	SC	Briquette	1.98	23.865	na ^a	na ^a	62.581
CX	MVB			2.51	3.791	0.065	0.779	108.633
ZY	MVB			1.92	3.594	0.068	0.756	63.001
CZ	LVB			0.19	2.429	0.071	0.498	46.863
AY	SA			0.07	0.720	0.031	0.246	59.157
YL	HVB	XF	Briquette	1.99	18.514	0.058	1.231	68.553
CX	MVB			1.01	6.815	na ^a	na ^a	91.587
ZY	MVB			1.54	6.055	0.035	1.161	64.870
CZ	LVB			0.25	2.670	0.011	0.288	87.161
AY	SA			0.08	0.526	0.004	0.222	75.366
		Average for briquette		1.154	6.898	0.043	0.648	72.777
		SD ^b		0.946	7.893	0.026	0.401	18.227

^a Data not available due to failed PAH measurement.

^b Standard deviation.

(Bonfanti et al., 1994; Chen et al., 2005, 2015; Shen et al., 2013; Zhi et al., 2008, 2009).

Two comparisons were designed based on the data presented in Table 1. The first compares PAH emissions between burning briquettes and chunks in the SC stove using the ratio of briquette to chunk emissions (Table 2, upper half). According to Table 2, the pPAHs, nPAHs, oPAHs, and aPAHs emitted from the burning of briquettes in the SC stove are 3.9 ± 2.9 , 7.5 ± 7.1 , 2.5 ± 1.2 , and 182 ± 241 times greater than those arising from the burning of coal chunks, respectively. These data highlight the dramatically elevated emissions of all 4 PAH groups due to the transformation of coal chunks to briquettes. Similar trends were also found for the EFs of OC in the same experiment (Chen et al., 2015), that 2.5 ± 1.1 times of OC emissions occurred after coal briquetting (Table 2), in contrast with the declined emissions of BC. For the second comparison, we calculated the ratio of average briquette emissions for the XF and SC stoves to the average chunk emissions for the GC and SC stoves (Table 2, lower half). The fold changes in PAH emissions due to briquetting are 3.1 ± 2.1 , 3.7 ± 0.7 , 1.9 ± 0.6 , and 171 ± 219 for pPAHs, nPAHs, oPAHs, and aPAHs, respectively. These data further support the finding of higher PAH emissions from burning coal as briquettes compared to chunks.

The increase in PAH emissions from burning briquettes would cause an increased health risk due to the toxicity and carcinogenicity of many PAHs (Luch, 2005; Mumford et al., 1987; Zhang and Smith, 2007), directly disputing the clean and healthy implications of coal briquetting. The health risk is further intensified by the extremely high increase in the aPAHs compared with the other PAHs. As shown in Table 2, briquetting increased aPAH emissions by approximately 180 times, which was significantly higher than the increases in the other 3 PAH groups (1.9–7.5). The aPAH group typically accounts for half of the total PAHs (including pPAHs and derivatives) and possesses stronger carcinogenicity and mutagenicity than pPAHs (Chen et al., 2005; Durant et al., 1996; Harvey, 1991). Thus, the distinctively higher aPAH emissions due to briquetting makes the increase in total PAH emissions an even greater human health threat.

3.2. Tentative insight into the increase

It is generally accepted that both PAHs and BC are products of incomplete combustion and are quite dependent on combustion efficiency. Our previous studies have proven that improving combustion efficiency by briquetting promotes the completeness of combustion and consequently decreases BC emissions (Chen et al., 2015; Zhi et al., 2008, 2009). However, the measured increase in PAH emissions from coal briquettes in the present study gave a strong indication that PAHs are not affected as much by combustion efficiency as BC and might be more greatly affected by other factor(s). After low- and high-ranking coals were burned in a fluidized bed reactor on a laboratory scale, Mastral et al. (1996) discovered that the total PAH emissions primarily depended on pyrolytic process instead of combustion efficiency. If this finding by Mastral et al. applies to household coal stoves, then the pyrolytic process rather than combustion efficiency would determine PAH emissions during residential coal burning. In this context it is critical to answer the following two questions: (i) Does coal briquetting facilitate pyrolytic process? (ii) Can the intensified pyrolysis lead to increased PAH emissions?

The answer to the first question seems to be 'Yes'. As formerly described, coal briquetting typically involves the pulverization of coal chunks by a coal crusher and the compression of the coal-clay-moisture mixture into a circular cylinder of 12–16 vertical holes by a briquette molder machine. The pulverization increases the surface area of the coal and the compression into a multi-hole cylinder

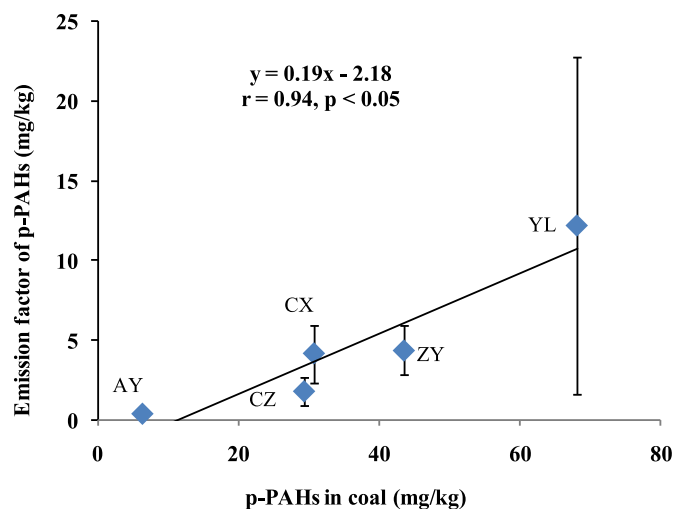


Fig. 1. The relationship between inherent pPAHs in raw coal and emitted particulate pPAHs. The sum of p-PAH emissions for each coal is an average over combinations of GC-chunk, SC-chunk, SC-briquette, and XF-briquette given in Table 1. Error bars represent the standard deviation of the mean.

enhances the introduction and circulation of supporting air; both of these actions facilitate an easy and close contact of the fuel with the surrounding high temperature air and thereby result in accelerated pyrolysis of the coal organics (although this is also accompanied by an increase in combustion efficiency) (Bond et al., 2002; Zhi et al., 2008, 2009).

The answer to the second question is relatively complicated but might be 'Yes' as well. PAHs emitted from coal combustion have two origins. One origin is the release of PAHs that are originally trapped in raw coals; the other is chemical formation by PAH precursors. For the first origin, high temperatures of combustion can liberate coal-trapped PAHs from the coal matrix (Li et al., 2003), and these liberated PAHs constitute an important part of the final PAH emissions. We extracted the PAHs from the five raw coals using DCM solvent and found that the extracted PAHs were in the range of 6.3–68.1 mg/kg (Table S3 in the SI), very close to the data reported by other studies (e.g., Huang et al., 2014). Furthermore, a significant correlation was observed between the particle-bound pPAHs emitted during coal combustion and the pPAHs contained in the raw coals (see Fig. 1 here and Table S4 in SI), which in some way supported the important role of trapped PAHs in their final emissions. Similar relationship can also be inferred from other studies involving PAH emissions from household coal combustion, though never has direct evidence been explored to support the mechanism (Chen et al., 2005; Huang et al., 2014; Shen et al., 2012).

For the second origin, PAHs can be formed from precursors via complex processes, for example, by chemical changes (Liu et al., 2009). Li et al. (2003) burned 4 steam coals in a self-designed tube furnace and found much more abundant PAHs in emissions than in the coal matrix, suggesting a significant contribution of chemically formed PAHs during coal combustion. This can be strengthened by the finding of Huang et al. (2014) that pPAHs, nPAHs, and oPAHs contained in raw coals only accounted for 5.17–39.9%, 1.21–12.1%, and 0.91–13.5% of their emission factors, respectively, in both particle and gas phases of coal smoke. In this study the EFs reported in the present study are only for the particle phase, they are usually lower than their contents in raw coals (Fig. 1). However, aPAHs emission factors for coal briquette combustion (72.8 ± 18.2 mg/kg in Table 1) are much higher than the levels of aPAHs contained in raw coals (27.2 ± 17.8 mg/kg in Table S3), directly confirming the existence of the second origin.

It is reported that the second origin (*de novo* formation of PAHs) is dependent upon two inter-linked processes: pyrolysis and pyrosynthesis (Barbella et al., 1990; Bjorseth and Ramdahl, 1985; Bonfanti et al., 1994; Mastral et al., 1996, 1999a, 1999b; Williams et al., 1986). Pyrolysis allows the organic compounds inside fuels to fall apart into smaller and less stable fragments, which can act as the precursors for subsequent pyrosynthesis (Flagan and Seinfeld, 1988) whereas pyrosynthesis converts these fragments of highly reactive free radicals into stable PAHs through recombination reactions such as the cyclization of alkyl chains and radical condensation (Mastral et al., 1996; Mastral and Callén, 2000).

3.3. Implications in research and policy

To our knowledge, we are the first to report a direct and systematic comparison of PAH emissions between chunk and briquette coal burning. The finding that burning coal briquettes leads to an increase in PAH emissions challenges the established notion that briquetting is a cleaner method compared with coal chunks. This finding makes us question the understanding and use of coal briquettes as CCT and their cobenefits to health and the climate and has important implications for framing future research and policy.

For future research, there are two issues calling for in-depth explanation. First, although we have found some evidence supporting the PAH enhancement by briquetting, given the diversity of coals, stoves, and burning conditions, and the complexity of PAH formation and emission, it is too early to reach a final conclusion based on a small dataset in such a single study. Therefore, various tests, not only laboratory experiments, but also field measurements, are needed to justify or deny the finding. Second, our assumption or hypothesis that pyrolytic process plays a decisive role in PAH EFs is only inferred from literature reports irrelevant with residential conditions, and the two questions raised in 3.2 were not confidently answered. Thus a special experiment needs to be designed to re-address the two questions by closely tracking the combustion-induced microscopic physical and chemical changes under household coal burning conditions.

When it comes to policy, we previously urged the government to support the substitution of coal briquettes for chunks to reduce the visible black smoke emissions (Zhi et al., 2009), which have been widely referred to by some influential journal studies and reports (e.g., Bond et al., 2013; Sasser et al., 2012; Shen et al., 2013; Stohl et al., 2013; UNEP and WMO, 2011). However, considering that China's newest clean air *Action Plan* attaches special importance to the contribution of coal to the worrisome haze across China's densely populated zones, and that both raw coal and briquettes are flawed either in their PM and BC emissions or in their PAH emissions, an ultimate ban on household coal use, accompanied by shift from coal to electricity or gas, together with subsidies to original coal users, seems to be a practical resolution of the problem in China. Globally, we notice that available important assessment reports in favor of coal briquette deployment failed to consider the possible increase in PAH or other pollutant emissions (Sasser et al., 2012; UNEP, 2011; UNEP and WMO, 2011; World Bank, 2013). Thus the agencies that propose coal briquetting need to append a caveat regarding PAH risks to their reports so that people can consider all possible options by choosing between health and cost.

4. Conclusion

In the present study, emission factors of various PAHs for household combustion of 20 coal/stove combinations were measured and compared in order to check the effects of coal

briquetting on PAH emissions. The EFs of 16 parent PAHs, 26 nitrated PAHs, 6 oxygenated PAHs, and 8 alkylated PAHs for coal briquettes were 6.90 ± 7.89 , 0.04 ± 0.03 , 0.65 ± 0.40 , and 72.78 ± 18.23 mg/kg, respectively, and were about 3.1, 3.7, 1.9, and 171 times higher than those for coal chunks, respectively. Such increase in PAH emissions due to coal briquetting contrasted to the significant decrease in some other pollutants (e.g., PM and BC), which may increase human health risk and challenge the policy of CCT. We assume that the pyrolytic process plays a critical role in PAH emission, followed by some preliminary explanations based on available information. However, further studies are required to find direct, rather than indirect evidence to justify and refine the assumed mechanisms of PAH emissions. Finally, we suggest that both China and the world should make a more comprehensive evaluation on coal briquette deployment if they hope to harvest climate-health cobenefit from coal briquetting.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2015.04.012>.

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