



Published in final edited form as:

Environ Sci Technol. 2013 March 19; 47(6): 2998–3005. doi:10.1021/es304599g.

Field Measurement of Emission factors of PM, EC, OC, Parent, Nitro- and Oxy- Polycyclic Aromatic Hydrocarbons for Residential Briquette, Coal Cake, and Wood in Rural Shanxi, China

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Abstract

Air pollutants from residential solid fuel combustion are attracting growing public concern. Field measured emission factors (EFs) of various air pollutants for solid fuels are close to the reality and urgently needed for better emission estimations. In this study, emission factors of particulate matter (PM), organic carbon (OC), elemental carbon (EC), and various polycyclic aromatic hydrocarbons (PAHs) from residential combustions of coal briquette, coal cake, and wood were measured in rural Heshun County, China. The measured EFs of PM, OC, and EC were 8.1–8.5, 2.2–3.6, 0.91–1.6 g/kg for the wood burnt in a simple metal stove, 0.54–0.64, 0.13–0.14, 0.040–0.0041 g/kg for the briquette burned in an improved stove with a chimney, and 3.2–8.5, 0.38–0.58, 0.022–0.052 g/kg for the homemade coal cake combusted in a brick stove with a flue, respectively. EFs of 28 parent PAHs, 4 oxygenated PAHs and 9 nitro-PAHs were 182–297, 7.8–10, 0.14–0.55 mg/kg for the wood, 14–16, 1.7–2.6, 0.64–0.83 mg/kg for the briquette, and 168–223, 4.7–9.5, 0.16–2.4 mg/kg for the coal cake, respectively. Emissions from the wood and coal cake combustions were much higher than those for the coal briquette, especially true for high molecular weight PAHs. Most EFs measured in the field were higher than those measured in stove combustions under laboratory conditions.

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The authors declare no competing financial interest.

Supporting Information

Supporting materials associated with this article are provided and available free of charge via the internet at <http://pubs.acs.org>.

Introduction

Biomass fuel and coal are extensively consumed all over the world. It was reported that in 2007, annual residential consumptions of coal and biomass fuels were approximately 3.34 and 36.5 EJ, respectively, of which more than 93% occurred in developing countries.¹⁻³ In China, residential consumptions of coal and biomass fuels were 2.1 and 8.4 EJ, respectively.¹⁻² Owing to low combustion efficiency, large amounts of air pollutants are generated from residential solid fuel combustions. The pollutants often concerned are CO, NO_x, SO₂, mercury, particulate matter (PM), black carbon (BC), polycyclic aromatic hydrocarbons (PAHs) including parent PAHs (pPAH), oxygenated PAHs (oPAHs), and nitrated PAHs (nPAHs).⁴⁻¹¹ It was estimated that residential solid fuel consumption contributed 82, 50, and 82% of pPAHs, BC, and primary organic carbon emissions in China, respectively.¹²⁻¹⁴

Globally, 35% of chronic obstructive pulmonary deaths, 21% of lower respiratory infections, and 3% of lung cancer deaths in 2004 were attributable to residential solid fuel combustions, causing a total number of 1.97 million deaths.¹⁵⁻¹⁶ In China, exposure to household solid fuel combustion was the largest single environmental risk, causing 0.42 million premature death annually.¹⁰ In addition to adverse health impacts, emissions from residential solid fuel combustion are also climate relevant. Carbonaceous particles from the residential sector are believed to contribute significantly to atmospheric brown cloud.¹⁷⁻¹⁸ It has also been suggested that the increased occurrence of summer floods in southern China might be partly explained by BC from residential solid fuel combustion.¹⁹

Emissions of air pollutants can be estimated based on emission factors (EFs) and fuel consumptions. Unfortunately, the measured EFs often vary several orders of magnitude and most EFs have been collected in developed countries to this stage, leading to high uncertainty in emission inventory.^{12-14,20-21} Significant differences in the measured EFs are often found between those measured in laboratory chambers and those measured in real kitchens, and between those under controlled conditions and those in field.²²⁻²⁵ Laboratory experiments allow investigators to repeat the process under controlled conditions for investigating the emission process, influencing factors, mechanisms, and kinetics.^{23-24,26-31} As such, laboratories are suitable for testing a variety of stoves operating under different conditions. However, direct field measurements are still preferable because they are more realistic and can cover emissions from stoves operated at random conditions that are difficult to reproduce in laboratory.³²⁻³⁶

In a previous study, EFs of carbonaceous PM and PAHs for residential solid fuels were measured using a typical brick stove for biomass and an iron moveable stove for coal in a rural kitchen.³⁷⁻⁴¹ The main objective of the present study was to conduct field measurements on EFs of PM, OC, EC, parent PAHs, oxygenated PAHs and nitrated PAHs (EF_{PM}, EF_{OC}, EF_{EC}, EF_{pPAH}, EF_{oPAH}, and EF_{nPAH}, respectively) for coal briquettes, homemade coal cake, and firewood. Typical cooking stoves often seen in rural China, including a movable metal stove for biomass, a brick stove and an improved metal stove for coal, were selected for the field test.

Experimental

Site Description

Field samples were collected in Heshun County in Shanxi, China (113°03′–113°56′ E, 37°03′–37°35′ N), where emission densities of PM and PAHs were much higher than the national average.^{13–14} Relatively high morbidities of a number of diseases including lung cancer and neural tube defects in this area have been reported.^{42–45} The annual net income per capita of the rural residents in 2011 was 3335 RMB,⁴⁶ much lower than the national (6977 RMB) and provincial averages (5601 RMB).^{47–48}

Like many other rural areas in China, various types of energy are used. Solid fuels, electricity, and liquid petroleum gas (LPG) are common for cooking, while solid fuels are also widely used for heating in cold seasons (about 5 months per year in this area). Of the solid fuels used, coal briquettes are quite popular, while wood and coal cake, a traditional homemade fuel, are also widely used. Honeycomb coal briquettes are commercially available while coal cake is always made at home by mixing raw coal with clay (2:1, v/v).

Field Sampling

The samples were collected in randomly selected households using wood, coal briquettes, or coal cake as cooking fuels during a period from May 5th to 14th, 2011. The samples were collected during cooking time of the residents, who were asked to operate normally. Wood was burned in a metal stove with a large and exposed firebox without chimney. Coal briquettes were combusted in an improved metal stove with an upper movable lid and a flue pipe. Coal cake was burned in a brick stove with an upper metallic grate and a chimney. Coals were all top-feed. During the sampling, the metal wood-burning stove and improved coal briquette stove were used for water boiling and the brick stove burning coal cake was used for cabbage frying. The pictures of tested stoves and fuels are shown in Figure S1, and measured fuel properties are listed Table S1.

Sampling probes were placed near the chimney center (for coal briquette and coal coke combustion) or 1.0 m above the stoves (for wood combustion).^{24,34–36} The exhaust smoke was naturally diluted by the ambient air during the initial several seconds.^{24–25} A total of 12 samples (3 fuels × 2 duplicate × 2 parallel PM filters) were collected and each sample was collected for 20 min.

PM samples were collected on quartz fiber filters (QFFs, 22 mm in diameter) using an active sampler at a flow rate of 1.5 L/min (XQC-15E, Tianyue, China). The flow rate was calibrated before and after each sampling period using a primary flow calibrator (Bios. Defender 510, USA). Two parallel PM samples were collected and used for EC/OC and particle-bound organic analysis, respectively. Gaseous PAHs and their derivatives were sampled on polyurethane foam plugs (PUFs, 22 mm diameter × 7.6 cm, density of 0.024 g/cm³). Before sampling, PUFs were pre-extracted with acetone, dichloromethane, and hexane in sequence (8 h for each). QFFs were pre-baked at 450°C for 6 h. Both PUFs and QFFs, before and after the sampling, were separately packed in clean aluminum foil. The samples were stored under –20°C before analysis.

CO and CO₂ concentrations in the exhaust were measured on site using an analyzer equipped with non-dispersive infrared sensors (GXH-3051, Tech. Inst., China). The instrument was calibrated using a span gas and zero-checked before and after each sample collection.

Analysis and Quality Control

The analysis procedures for PM, EC, OC, pPAHs, oPAHs, and nPAHs were the same as those in previous studies.^{38,41} Briefly, PM gravimetric analysis was conducted using a high precision digital balance (0.01 mg). EC and OC were measured offline using a Sunset semi-continuous EC/OC analyzer (RT-4, Sunset, USA). The temperature protocol is provided in the Supporting Information (Table S2). Pyrolyzed organic carbon was determined when the transmitted light intensity returned back to the initial value, and subtracted from the EC result. Failure to calibrate the oven temperature may cause problems in the split of EC and OC in transmittance correction,⁴⁹ although it was reported that when the temperatures of filter and oven differed, there were statistically significant differences in the sub-carbon fractions, but insignificant differences for the total OC, EC and total carbon content.^{50–51}

For PAHs, the PUFs were Soxhlet extracted with 150 mL dichloromethane for 8 h. QFFs were extracted with 25 mL hexane/acetone mixture (1:1, v/v) using a microwave accelerated system (CEM Mars Xpress, USA, 1200 W) operated at 1200 w (100%). The temperature was increased to 110 °C in 10 min and then held for another 10 min. Silica/alumina gel columns were used for cleanup. The column was eluted with 20 mL hexane (discharged) and then 70 mL hexane/dichloromethane mixture (1:1, v/v), which was collected, concentrated to 1 mL, spiked with deuterated internal standards (J&W Chemical, USA) and finally analyzed using a gas chromatograph – mass spectrometer (Agilent, USA) equipped with a HP-5MS capillary column. Electron ionization and negative chemical ionization modes were used for the analysis of pPAHs and their derivatives, respectively. Compounds were identified based on the retention time and qualitative ions of the standards in selected ion mode. Twenty-eight pPAHs, four oPAHs, and nine nPAHs were analyzed in the current study (Table S3). Detection limits and recoveries of spiked standards for the targets are provided in the Table S3. Recoveries of the surrogates (2-fluoro-1,1'-biphenyl and *p*-terphenyl-*d*₁₄, J&W Chemical, USA) added to monitor the laboratory analysis were 76–105 and 92–120% for gaseous samples, and 88–100 and 95–117% for particulate samples, respectively. Blanks were measured and subtracted from the sample measurements.

Data Analysis

EFs were determined using the carbon mass balance method, which assumes that carbon in the fuel emits in the forms of gaseous CO, CO₂, total hydrocarbon, and carbon in PM during combustion.^{32,52–53} It was reported that omission of total hydrocarbon in gas would not cause a large error.³² The detailed calculation process can be found elsewhere.^{37,52} Modified combustion efficiency (MCE), defined as CO₂/(CO+CO₂) (molar basis) is calculated to characterize the combustion. Since most carbon is emitted as gaseous CO and CO₂, the difference between MCE and combustion efficiency, defined as carbon in CO₂ divided by the total carbon released, is very small.^{54–55} Statistica (v5.5, StatSoft) was used for data statistical analysis with a significance level of 0.05.

Results and Discussion

The measured EFs are summarized in Table 1 in terms of pollutant mass per burned fuel mass (g/kg). EFs of OC and EC reported here are the particle carbon contents and EFs for parent PAHs and their derivatives are sums of gaseous and particulate phases. In addition to a total EF of 28 pPAHs (EF_{28pPAH}), a total EF of 16 priority PAHs (EF_{16pPAH}) is also provided. EFs of individual pPAH, oPAH, and nPAH are listed in Table S4.

Wood Combustion

In a previous field study conducted in China, it was reported that the overall means of EFs of $PM_{2.5}$ (PM with diameter less than 2.5 μm), OC, and EC for wood were 3.1 ± 0.82 , 1.14 ± 0.40 , and 1.5 ± 0.69 g/kg, respectively.⁵⁶ Table S5 lists reported EF values for residential wood combustion measured in rural China. So far, most EFs were measured in laboratory conditions. It is interesting to see that the EFs reported based on laboratory tests were generally lower than those obtained in the field. Zhang *et al.* reported an EF_{PM} of 1.5–4.1 g/kg for wood burned in different stoves.⁵² Recent measurements of EFs for residential wood combustion in a typical cooking stove yielded EF_{PM} , EF_{OC} , and EF_{EC} values of 1.6 ± 0.32 , 0.60 ± 0.35 , and 0.94 ± 0.40 g/kg, respectively,³⁸ which were 5.2, 4.8, and 1.3 times lower than the results obtained in this field test. Based on a field measurement in Honduras, it was also reported that EF_{PM} values for wood burned in residential stoves were 2–4 times higher than those in laboratory tests.²⁵ The stove used in our previous experiment³⁸ was a so-called “improved” one with a shorter distance between the grid and cookware, a smaller firebox, and a taller chimney. The improved stoves are now commonly used in many places in China after the implementation of the National Improved Stove Program during the 1980s to 1990s.⁵⁷ However, the simple movable wood stove without flue tested in this study (Figure S1) is also used quite often.^{9,58} It is generally expected that the combustion efficiency in a stove with a chimney is higher than that without one due to an increased draft condition.^{59–60} In addition, mainly wood branches, instead of wood logs, were burned. This could also lead to relatively high emissions, since EFs for brushwood/branch were reported to be higher than those for fuel wood logs.^{9,38} Another possible reason for higher emissions is that the fire management behavior in the laboratory test was essentially “normalized”, while the process observed in the field was rather random, which could include some low combustion efficiency occasions.

The ratios of EC/OC, EC/PM, and OC/PM are often used as important indicators for source apportionment and climate impact analysis of carbonaceous PM.^{61–63} Calculated ratios of EC/OC, EC/PM, and OC/PM from the wood combustion were 0.42 ± 0.02 , 15 ± 6 , and $35 \pm 13\%$, respectively. In comparison with the results of the previous laboratory test (2.1 ± 1.3 , 49 ± 20 , and $31 \pm 16\%$),³⁸ it appears that a higher proportion of EC, but not OC, was generated while operating the improved stove with a chimney under laboratory conditions. Such a discrepancy can be explained again by the fact that under laboratory conditions, the stove performed better than that in the field test, and the former test may fail to reproduce some low-efficiency burning occasions.²⁴ In fact, these ratios varied dramatically not only among sources, but also among burning conditions,^{5,63} suggesting that caution should be exercised when using these ratios. The EFs of PAHs for residential wood combustion were

tested in the field for the first time in this study. The measured EF_{28pPAH} and EF_{16pPAH} were about two orders of magnitude higher than those detected in the previous combustion experiments using an improved stove (6.4 ± 2.8 and 6.0 ± 2.7 mg/kg, respectively).⁴¹ The reasons for the relatively high EF_{pPAH} are similar to those for PM. However, the difference in EF_{pPAH} was larger than that in EF_{PM} . A larger difference in EF_{pPAH} between two distinct combustion emissions in comparison with EF_{PM} was also mentioned in the literature,^{22,64} indicating that the formation and emission of PAHs could be more sensitive to the variability in fuel/stove types and fire management behavior than PM.

Though EF values for pPAHs measured in field were higher than those from the laboratory tests, the composition profiles were similar to each other, made up of approximately one third of NAP, followed by FLA, PYR, and PHE. Attention should be paid to those compounds with relatively high molecular weight PAHs (> 228) that are often carcinogenic.^{65–67} The 7 carcinogenic compounds with high molecular weight (BaA, CHR, BbF, BkF, BaP, DahA, and IcdP) accounted for 16% of the total emissions of the 16 U.S. EPA priority PAHs. This percentage was much higher than 6.3% of the global total emissions from all sources.⁴³ Similar to the 16 priority pollutants, EFs for 12 other parent PAHs, most of which were highly toxic,^{65,68} were also much higher than those measured in the previous combustion under laboratory conditions. For example, EFs of CPP and DalP in the present field measurement were 17 ± 7.0 and 0.46 ± 0.14 mg/kg, while those from the combustion under laboratory conditions were only 0.021 ± 0.0030 and 0.0020 ± 0.0018 mg/kg, respectively.⁴¹

EFs of oPAHs and nPAHs were also measured for the first time in the field. For the same reason discussed above, these EFs were 1–2 orders of magnitude higher than the values reported in the previous laboratory combustion test (0.37 ± 0.27 and 0.0051 ± 0.0029 mg/kg for EF_{40PAH} and EF_{9nPAH} , respectively).⁴¹ Of the 4 oxy-PAHs measured, EFs of 9FO (3.5 mg/kg) and BZO (3.7 mg/kg) were the highest, followed by ATQ (1.7 mg/kg). EFs of 9FO and ATQ were the same order of magnitude as those of corresponding parent PAHs and EF of BaAQ was around 2 orders of magnitude lower than that of BaA. The calculated EF ratios of oxy-PAHs over their corresponding parent PAHs were 0.79 (9FO/FLO), 0.42 (ATQ/ANT), and 0.020 (BaAQ/BaA), which were close to those in emissions from the previous laboratory test.⁴¹ Among all nitro-PAHs measured, 9N-ANT and 9N-PHE were the two most abundant species having EFs of 0.047–0.33 and 0.042–0.18 mg/kg, respectively. Similar to the results derived previously in the combustion under laboratory conditions,⁴¹ EFs of individual nPAHs were 2–5 orders of magnitude lower than EFs of their corresponding pPAHs. The calculated EF ratios of nPAHs over their corresponding pPAHs ranged from 0.000057 (EF_{1N-NAP}/EF_{NAP}) to 0.095 (EF_{9N-ANT}/EF_{ANT}).

Coal Briquette Combustion

To the best of our knowledge, field measurements of EFs of residential coal briquette combustion in rural China have not been conducted previously. The ratios of EC/OC, EC/PM, and OC/PM for the coal briquette were 0.031 ± 0.002 , $0.69\pm 0.07\%$ and $22\pm 4\%$, respectively. These measured EFs were high in general in comparison with those measured from combustions under laboratory conditions. For instance, EF_{PM} , EF_{OC} , and EF_{EC} of the

coal briquettes (also from Shanxi) that were burned in an iron stove were 0.17 ± 0.010 , 0.021 ± 0.0020 , and 0.0042 ± 0.0021 g/kg, respectively.³⁷ It was also reported that EF_{PM} of honeycomb briquettes burned in residential metal stoves were in the range of 0.032 to 0.62 g/kg.⁵² Coal properties including chemical compositions, volatile matter, and ash contents often vary widely, resulting in differences in combustion characteristics and emissions of various incomplete combustion byproducts.^{21,62,69–71} For example, EF_{PM} , EF_{OC} , and EF_{EC} were reported to be 1.3, 0.017, and 0.0040 g/kg for anthracite briquettes and 7.8–20, 3.6–14, and 0.064–0.68 g/kg for bituminous briquettes, respectively.⁶⁹ Therefore, the differences in EFs between this study and others could be caused by the varying properties of coal from which the briquettes were made of. In addition, stove type and configuration are also critical in emission measurements. It was estimated that the emissions of PM, OC, and EC for coals burned in improved stoves with an upper lid and chimney could be reduced by 56, 61, and 14% compared to emissions from a traditional stove with a lid near the bottom and without a chimney.⁶²

For PAHs and their derivatives, measured EFs are again much higher than those measured in the laboratory experiment of coal briquette combustion (14 ± 3.1 and $2.9\pm 0.16\times 10^{-1}$ mg/kg for EF_{16pPAH} and EF_{40PAH} , respectively, while nitro-PAHs were not quantified).^{39–40} Low and median molecular weight PAHs, including PHE ($22\pm 0.11\%$), NAP ($18\pm 2.9\%$), FLA ($16\pm 1.3\%$), and PYR ($12\pm 0.91\%$) dominated the pPAH composition profile from the briquette combustion. Of the 4 oPAHs, EFs of 9FO ($1.1–1.8$ mg/kg) and ATQ ($0.50–0.68$ mg/kg) were higher than the other two oPAHs. Similar to emissions from the wood burning, EF_{9FO} and EF_{ATQ} were of the same order of magnitude as EFs of their corresponding parent PAHs, while EF_{BaAQ} was roughly 2 orders of magnitude lower than EF_{BaA} . The ratios of 9FO/FLO, ATQ/ANT, and BaAQ/BaA were 4.3, 1.7, and 0.025, respectively. Also like the wood fuel combustion, EF_{nPAH} values were 2–5 orders of magnitude lower than those of their corresponding parent PAHs with a single exception of 9N-ANT, whose EF was 0.48–0.59 mg/kg, comparable to its parent ANT ($0.33–0.38$ mg/kg).

Coal Cake Burning

Of the total PM mass emitted from the coal cake burning, OC and EC fractions were 9.3 ± 3.6 and $0.65\pm 0.06\%$, respectively. The calculated EC/OC ratio was 0.075 ± 0.022 . There was only one previous study in the literature reporting the emissions of PM and BaP from a similar coal cake made of raw coals from rural Shanxi.⁷² It was reported that EF_{PM} was 1.0 ± 0.06 g/kg, and EF_{BaP} was 2.1 ± 2.2 μ g/kg, respectively.⁷² These EFs were much lower than what we found in the present study. Different types of coals that were used in the made of coal cake, in addition to different stove designs, may explain the difference between the two studies. The volatile matter and ash contents of the coal cake in our study were 6 and 87%, respectively, while they were 11 and 30% of the coal cake tested by Ge *et al.*⁷²

For parent PAHs, low and median molecular weight compounds again dominated in the emissions. Approximately 25 ± 9.8 , 19 ± 0.3 , and $14\pm 3.7\%$ of the total 28 PAHs were NAP, ACY, and PHE. Of the 4 oPAHs from the coal cake burning, the EF of 9FO was the highest. The calculated ratios of 9FO/FLO, ATQ/ANT, and BaAQ/BaA were 0.39, 0.12, and 0.007,

respectively. Nitro-PAHs tested were again orders of magnitude lower than those of their corresponding parent PAHs. 9N-ANT and 9N-PHE were the most abundant compounds.

Fuel Comparison

EFs for the target pollutants were significantly different ($p < 0.05$) among the fuels, except for EF_{9nPAH} ($p = 0.346$). The highest values were found for the burning of wood, and the lowest for the coal briquette. The EF_{28pPAH} of the coal briquette was more than an order of magnitude lower than those of the coal cake and wood. The difference in MCE may be one reason for different EFs among the three fuels. The measured MCE for the wood ($92 \pm 3\%$) and the coal cake ($86 \pm 10\%$) combustions were significantly lower ($p = 1.3 \times 10^{-3}$ and 1.0×10^{-4} , respectively) than that of the coal briquette combustion ($94 \pm 4\%$). The effect of MCE has been documented in the literature. It was generally recognized that the lower the MCE, the higher the emissions of various incomplete combustion by-products.^{53–55} It was previously demonstrated that the measured EFs of PM and pPAHs can be predicted based on several factors including MCE, moisture, and volatile matter contents, and among these predictors, MCE is the only one that was significant across fuel types.^{37,39–40}

Coal cake is often thought as a very dirty fuel causing high emissions of various air pollutants due to low burning efficiency.^{44,72} EF_{PM} , EF_{OC} , EF_{EC} , EF_{28pPAH} , EF_{40PAH} , and EF_{9nPAH} for the coal cake were 9.9, 3.6, 9.2, 14, 3.3, and 1.8 times the associated EF values for the coal briquette, respectively. In addition to the difference in MCE, ash content of the coal cake (87%) was much higher than that of the coal briquette (47%). It was well established that ash content can affect the fuel pyrolysis process and prevent the oxidation reaction of compounds during the combustion.^{73–75} Moreover, an increase in ash content can also reduce the heating values of coal.^{40,75} Therefore, high ash content may lead to higher emissions. In addition, incombustible ash content can remain in the exhaust smoke resulting in higher particle emissions.²¹ High emissions of other pollutants such as SO_2 and NO_x from coal cake combustion in comparison with coal briquette combustion were also reported.⁷² Stove design, fuel addition interval and fire management practice can surely affect the measured emissions. Unfortunately, it is very difficult to quantify these factors in practice.

Wood fuel is extensively used in rural China and many other developing countries. In this study, the EFs for the wood were found to be higher or equivalent to those for the coal cake. This is particularly true for OC and EC. EF_{OC} and EF_{EC} for the wood burning were 6.4 and 40 times of those for the coal cake, and 22 and 306 times of those for the coal briquette. Higher emissions for wood can be partly explained by relatively low combustion efficiency and high volatile matter content (Table S1). Although overall average MCE for the wood was close to that for the briquette, the difference was statistically significant, most likely because EFs measured in this study are filter based, rather than real time data. Future studies focusing on the real time emissions are needed for a full understanding.

It was shown for all the three fuels that EF_{9FO} and EF_{ATQ} were comparable to their parent PAHs of FLO and ANT (within 0–1 order of magnitude difference), and EF_{BaAQ} and EF_{nPAHs} were about 2–5 orders of magnitude lower than EFs of their corresponding parent PAHs. Although the total emissions of pPAHs varied dramatically among the three fuels, the

composition profiles were similar in general (Figure 1). For all three fuels tested, the parent PAHs were dominated by low and median molecular weight compounds especially NAP, ACY, PHE, and PHE. The only noted differences were the relatively high fractions of CPP, BaP, and PAHs with molecular weight larger than 302^{65,68} from the wood and coal cake combustions. For instance, of the total 28 PAHs, mass percentages of CPP, BaP, and DaP were 5.3, 1.6, and 0.26% in the coal cake emission, and 8.1, 2.3, and 0.21% for the wood combustion, but only 0.48, 0.50, and 0.041% in the emission from the coal briquette combustion. It is revealed that the coal cake and wood combustion cause not only higher emissions, but also larger fractions of highly toxic PAHs. Exposure to these highly toxic PAHs, in either indoor or outdoor environments, is believed to be associated with high risks of many diseases, including lung cancer.^{43–44,76} The composition profiles of PAH derivatives for all individual tests are shown in Figure S2. Again, the normalized profiles for all three tested fuels were similar although EFs varied significantly. Generally, 9FO and BZO were two oxy-PAHs with higher emissions, and 9N-ANT and 9N-PHE were the two highest emitted nitrated PAHs. However, since only 4 oxy-PAHs and 9 nitrated PAHs were measured in this study, the information should be explained or used with care.

It has been reported previously that the emissions of PMs and PAHs from stoves in real kitchens are higher than those measured using chambers.^{7,24,37,69} In this study, it was found that the field measured emissions of various pollutants from the wood and coal combustions were higher than (or at least different from) those reported in stove combustions conducted in a laboratory under controlled conditions. It appears that the latter can cause underestimation of the emissions. In laboratory studies, new stoves purchased directly from market are often used. The combustion efficiency is expected to go down as the stoves wear out and the flue gets choked. It was reported that after a stove was used for about one year, EF_{PM} increased by approximately 50%.²⁵ Another reason for the difference between laboratory and field tests is the way of refuel. In a laboratory, the refueling process is unintentionally normalized by the researchers, who are trained to reproduce the experimental results, while in the reality, however, fuels are often introduced randomly. Therefore, more realistic EFs, with high variations, can only be obtained in a field survey, while laboratory tests are useful for understanding the emission processes and influencing factors and conditions.

EFs for different fuels can be very different. In this study, the EFs of most pollutants from the wood and coal cake burning were orders of magnitude higher than those of the coal briquette. Both wood and coal cake are extensively used in rural China,^{9,72} as well as in many other developing countries. There is an enormous potential to reduce air pollutant emissions in rural areas of these countries by replacing wood and coal cake (also chunk coal) with cleaner fuels, taking into consideration the large differences in the EFs of many air pollutants. In fact, coal briquette usage in households in China has been growing.¹⁴ Use of compressed biomass can also help to reduce harmful emissions. It was reported that EFs of carbonaceous PM for wood pellets are significantly lower than corresponding values for wood fuel.⁷⁷ It is clear that the replacement of solid fuels with cleaner energy can help to improve ambient and indoor air qualities, and subsequently reduce adverse health impacts.⁷⁸

It should be noted that this study mainly focused on specific fuel/stove combinations in rural Shanxi, and included rather limited sample collections due to high labor intensity, cost, and technical difficulty. Therefore, the results of this study cannot be generalized. Instead, the results provide us valuable data and help further understanding of the difference between field and laboratory tests. It is also realized that EFs reported in the literature often vary in orders of magnitude, so the difference within one order of magnitude between the field tests and laboratory measurements found in the current study is relatively small. It is reasonable to expect that variation in the field-measured EFs should be much higher than that measured in laboratory tests. More campaigns with relatively large sample sizes are strongly recommended before emissions can be fully understood.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

Funding for this study was provided by the National Natural Science Foundation of China (41001343, 41130754, and 41101490), Beijing Municipal Government (YB20101000101), and NIEHS (P42 ES016465). The authors thank Li Zhao from Maternal and Children Health Care Service Center in Heshun County and Yangyang Song, Li Zhao and Beibei Song from Shanxi Agricultural University, and the rural residents who helped us in sample collection. We thank Kelly McCusker from University of Washington for polishing the language.

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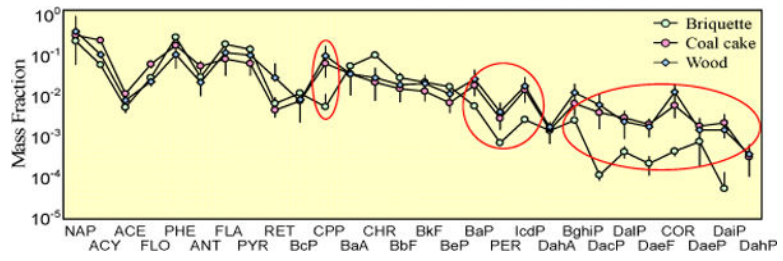


Figure 1. Comparison profiles of the measured parent PAHs for the wood, coal cake and coal briquette measured in this study.

Table 1

EFs of PM, OC, EC, total parent PAHs, oxy- and nitro-PAHs from residential combustions of coal briquette, coal cake, and wood. Data shown are the range from duplicate measurements. Reported values for PAHs and their derivatives are gas- and particle-phase are combined.

	Briquette	Coal Cake	Wood
PM , g/kg	0.54–0.64	3.2–8.5	8.1–8.5
OC , g/kg	0.13–0.14	0.38–0.58	2.2–3.6
EC , g/kg	0.0040–0.0041	0.022–0.052	0.91–1.6
28 pPAHs , mg/kg	14–16	168–223	182–297
16 pPAHs , mg/kg	13–15	148–210	141–276
4 oPAHs , mg/kg	1.7–2.6	4.7–9.5	7.8–10
9 nPAHs , mg/kg	0.64–0.83	0.16–2.4	0.14–0.55