



## Field based measurement of multiple pollutant emissions from residential coal burning in rural Shanxi, northern China

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### ARTICLE INFO

#### Keywords:

Residential coal combustion  
Emission factors  
Particulate matter  
Polycyclic aromatic hydrocarbons

### ABSTRACT

It was well recognized that the pollutant emissions from real-world residential coal combustion contributed significantly to air pollution and human health, resulting in an urgent need for a better understanding of the pollutant emission factors (EFs) of residential coal combustion. However, field-based researches on residential coal combustion emissions are still limited at this stage. In this study, based on carbon balance method, emission factors (EFs) of CO<sub>2</sub> and a series of pollutants, including gaseous pollutants (CO, NO<sub>x</sub>, CH<sub>4</sub>, and SO<sub>2</sub>), particulate matters (PM<sub>2.5</sub> and PM<sub>10</sub>), elemental carbon (EC), organic carbon (OC), and polycyclic aromatic hydrocarbons (PAHs) from residential honeycomb briquette (HB) and coal chunk (CC) combustions were measured in real-world cooking stoves. As a result, apart from SO<sub>2</sub>, the burning of HB produced much less pollutants than CC. The median of emission reduction ratios estimated from Monte Carlo simulation ranged from 2.4% (SO<sub>2</sub>) to 98.8% (EC) by replacing CC with HB in residential combustion in Shanxi province in 2012. EFs of CO, PM<sub>2.5</sub> and PAHs derived from the present field measurement were around 1–50 times higher than those from laboratory tests; furthermore, larger variations were found in our field measurement compared to that in the laboratory studies. This result called for more field measurements to reduce uncertainties of emission inventory estimations. As for the PAH emissions, particle phase PAHs dominated in both HB and CC burning and PAH emissions from HB burning had lower toxicity than CC combustion. Hence, HB is an ideal substituted fuel compared with CC. Previous studies suggested that PAH isomer ratios of FLA/(FLA+PYR) and IcdP/(IcdP+BghiP) above 0.5 represented source of coal combustion, which did not fit with our study well, indicating PAH isomer ratios might not be valid enough for source apportionment.

### 1. Introduction

In China, there are still enormous rural population rely on coal for heating and cooking (Du et al., 2018a; Duan et al., 2014; Tao et al., 2018), especially in Shanxi province, attributed by easy access to coal, this proportions could be as high as 81.2% and 58.6%, respectively. Due to relatively low burning efficiencies of residential cooking stoves, the incomplete combustion of coal produced large quantities of various pollutants. For example, it was reported that residential coal burning

contributed to 13%, 22%, 23% and 36.5% of the total organic carbon (OC), elemental carbon (EC), polycyclic aromatic hydrocarbons (PAHs), and particles with aerodynamic diameter less than 2.5 μm (PM<sub>2.5</sub>) emissions in China, respectively (Lu et al., 2011; Zhang et al., 2007, 2018; Li et al., 2016a). Moreover, the widely used of coal for indoor burning resulted in not only the severe indoor and ambient air pollution, but also serious threats on human health (Chen et al., 2016; Du et al., 2017a; Wang et al., 2018). For example, it was reported that in rural China, indoor particles with aerodynamic diameter less than 10 μm

Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control.

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<https://doi.org/10.1016/j.apr.2020.12.005>

Received 2 July 2020; Received in revised form 8 December 2020; Accepted 9 December 2020

Available online 30 December 2020

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(PM<sub>10</sub>) and benzo[a]pyrene (BaP) concentrations could be  $420 \pm 30 \mu\text{g}/\text{m}^3$  and  $29.6 \pm 22.7 \text{ ng}/\text{m}^3$  when coal was burned in household (Fan et al., 2012; Wu et al., 2015), way above Chinese Air Quality Standards for indoor air, which were  $150 \mu\text{g}/\text{m}^3$  and  $1.0 \text{ ng}/\text{m}^3$  (MEP, 2002), respectively. According to the estimation by Zhao et al. (2018), 43% of premature death caused by air pollution in China occurs in rural areas, where people exposed to PM<sub>2.5</sub> emitted from solid fuel combustion for a long period, especially in winter.

To quantify pollutant emissions and estimate the impact of pollution from residential coal burning on the environment and human health, emission factors (EFs) with high quality, which could be used to calculate pollutant emissions together with fuel consumption amounts, were imperatively needed. Pollutant EFs can be measured under laboratory conditions or real-world conditions, which may vary greatly due to different emission patterns (Du et al., 2020a). However, most previous studies were conducted in laboratories or simulated kitchens under well controlled conditions (Chen et al., 2005, 2006; Shen et al., 2010a, 2010b), field measurements on emissions from residential stoves were very scarce to date (Du et al., 2020a; Shen et al., 2015), owing to high costs and the difficulty in field sampling. Due to various influencing factors such as air supply, operation management and stove types, EFs from field combustion could differ from those measured in laboratories under well-controlled conditions (Shen et al., 2011, 2013a, 2013b). Considering it is generally accepted that field-based tests can provide more realistic EFs than laboratory tests, the data gap in field tests was an important uncertainty source when emission inventories compiled in China (Wei et al., 2014). Hence, there is an urgent necessity for more field tests to obtain more realistic pollutant EFs.

In this study, field measurement was conducted in rural Shanxi province, where residents were suffered from severe indoor air pollution caused by residential coal combustion (Du et al., 2017b). This field measurement was expected to: 1) measure EFs of CO<sub>2</sub> and multiple pollutants including CO, NO<sub>x</sub>, CH<sub>4</sub>, SO<sub>2</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>, OC, EC and PAHs (EF<sub>CO2</sub>, EF<sub>CO</sub>, EF<sub>NOx</sub>, EF<sub>CH4</sub>, EF<sub>SO2</sub>, EF<sub>PM2.5</sub>, EF<sub>PM10</sub>, EF<sub>OC</sub>, EF<sub>EC</sub>, EF<sub>PAHs</sub>) from the burning of honeycomb briquettes (HB) and coal chunks (CC) in residential stoves; 2) compare EFs derived from field tests with those determined in laboratory; and 3) analyze composition profiles, isomer ratios, and gas-particle partition of PAHs emitted from residential coal combustion.

## 2. Materials and methods

### 2.1. Emission measurement

This study was conducted in Taigu country, Shanxi province located in northern China. HB and CC are two commonly used solid fuels for cooking in this area. A total of 20 households were selected randomly for emission tests, half of which used HB and the other half used CC. EFs of multiple pollutants emitted from residential coal combustion were measured when the residents cooked their lunch or dinner. According to our surveys, CC were produced in local coal mine, while HB were produced by compressing pulverized coal with clay and additives (i.e. NaNO<sub>3</sub>, KNO<sub>3</sub>, CaO) by individual workshops. Elemental contents including C, H, N and S were determined by elemental analyzer (Vario EL cube, Elementar Analysensysteme GmbH, Germany) and moisture was determined by halogen moisture analyzer (XY50MW, Xingyun Electronic Equipment, China), results were showed in Table S1. In addition, the sampling system in this study was provided in Fig. S1.

The tests were conducted during regular cooking time, residents were asked to operate stoves as they normally did. The HB and CC were burned in iron stoves equipped with an outdoor chimney. During sampling processes, the sampling probe was placed at the center of the chimney exit, smoke was extracted into a mixing room through sampling pump (AirChek XR 5000, SKC, USA). CO, CO<sub>2</sub>, CH<sub>4</sub> were measured using a nondispersive infrared sensor (GXH-3051, China), while NO<sub>x</sub> and SO<sub>2</sub> were measured by electrochemical and infrared sensor (JFQ-3150E,

China). PM<sub>2.5</sub>, PM<sub>10</sub> and gaseous PAHs were collected by glass fiber filters (GFFs, 37 mm in diameter) and polyurethane foams (PUFs, 22 mm in diameter and 7.6 cm in length, density of 0.024 g/cm<sup>3</sup>), respectively. To eliminate background interference, PUFs were pre-soxhelt-extracted in the sequence of acetone, dichloromethane and n-hexane, GFFs were prebaked at 450 °C, for 6 h in a muff furnace prior to the use. Before and after each sampling, all pump flows were calibrated using a flow calibrator (Bios Defender 510, USA). Each sampling process lasted for about 30 min.

### 2.2. Laboratory analysis

The particles were weighed using an electronic balance (0.01 mg, XS105, Mettler Toledo, Switzerland), filters were equilibrated for 48 h in a laboratory desiccator before weighing. An EC/OC analyzer (Sunset Lab, USA) was used to determine EC and OC in PM<sub>2.5</sub>. The filters were placed in pure helium and followed temperature protocols of 600, 840, and 500 °C to quantify OC, then heated at 550, 650, 500 °C in the mixture of oxygen and helium for EC detection (Du et al., 2018a).

The PAHs analysis procedures followed previous studies (Shen et al., 2010a; Du et al., 2020b). Briefly, Soxhlet extraction was used to extract the PUFs with 150 mL the mixture of n-hexane/acetone (1:1, V/V) for 8 h. While the GFFs were analyzed using a microwave extraction system (CEM, Mars Xpress, USA) extracted with 25 mL n-hexane/acetone (1:1, V/V), at the rate of 10 °C/min heated to 110 °C, then held for another 10 min. After extraction, using a rotary evaporator (N-1100; EYELA, Bunkyo-ku Tokyo, Japan) concentrated the extracts to about 1 mL, added 5 mL n-hexane and concentrated to 1 mL again, then transferred to a Silica/alumina gel column for cleanup. 20 mL hexane was used to pre-elute column, and then eluted with 70 mL the mixture of hexane/dichloromethane (1:1, V/V). The eluate was concentrated to 1 mL, added 5 mL n-hexane and concentrated to 1 mL at last, changed to hexane solution, and spiked with 200 ng deuterated standards.

PAHs were analyzed by a gas chromatograph coupled with mass spectrometer (GC-MS, Agilent GC 6890, MS 5973, USA) in electron ionization mode, equipped with HP-5MS capillary column (30 m × 0.25 mm × 0.25 μm, J&K Chemical, USA). The oven temperature protocol was 50 °C held for 1 min, 10 °C/min heated to 150 °C, 3 °C/min increased to 240 °C, and finally to 280 °C held for another 20 min. PAHs were identified and quantified based on the retention time and qualitative ions of the standards in selected ion mode and the use of selected quantitative ions for each compound, respectively. 28 PAHs including 16 U.S. EPA priority PAHs: naphthalene (NAP), acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), BaP, dibenzo[a,h]anthracene (DahA), indeno[1,2,3-cd]pyrene (IcdP), benzo[g,h,i]perylene (BghiP), and 12 non-priority parent PAHs: benzo[c]phenanthrene (BcP), retene (RET), perylene (PER), benzo[e]pyrene (BeP), coronene (COR), dibenzo[a,e]fluoranthene (DBaEF), cyclopenta[c,d]pyrene (CcdP), anthanthrene (AA), dibenzo[a,i]pyrene (DBaiP), dibenzo[a,l]pyrene (DBalP), dibenzo[a,e]pyrene (DBaEP), and dibenzo[a,h]pyrene (DBahP) were measured.

### 2.3. Quality control and data analysis

Blanks of GFFs and PUFs were determined and subtracted from the sample results. For quality control, 20% samples were selected randomly and added with two surrogates (2-fluoro-1, 1'-biphenyl and p-terphenylid14, Accustandard Inc., via J&W Scientific, USA). For the PUFs samples, the recoveries of surrogates were in the range from 66% to 105% and from 76% to 136%, respectively, while for GFFs samples, they were 80%–131% and 72%–110%, respectively. In addition, the instrument detection limit (IDL) of target pollutants, method detection limit (MDL) and recoveries of individual PAHs were presented in Table S2.

The pollutant EFs were calculated on the basis of carbon mass bal-

ance method, which was widely used in field measurements (Shen et al., 2010a). Monte Carlo simulation (10,000 replications) was implemented to evaluate the uncertainties of emission estimation based on EFs and fuel consumptions from the given distribution and the results were showed as median and interquartile range. The EFs followed log-normal distribution in this study, as for fuel consumption data, uniform distribution with a variation of 20% was suggested by Shen et al. (2015). Modified combustion efficiency (MCE, defined as  $\text{CO}_2/(\text{CO} + \text{CO}_2)$ ) was used as a proxy of combustion efficiency. The BaPeq (BaP equivalent EFs) was calculated to indicate the toxicity of PAHs (Du et al., 2018b), equation (1) was:

$$\text{BaPeq} = \sum_{i=1}^{n=28} \text{TEF}_{\text{PAH}_i} \times \text{EF}_{\text{PAH}_i} \quad (1)$$

where  $\text{TEF}_{\text{PAH}_i}$  was the toxicity equivalency factor (TEF) of individual PAH compound  $i$ ;  $\text{EF}_{\text{PAH}_i}$  was EF of individual PAH compound  $i$  (mg/kg) (see Table S2). The 16 priority PAHs and the 28 parent PAHs determined were denoted as  $\text{PAH}_{16}$  and  $\text{PAH}_{28}$ , respectively.

SPSS 21.0 (IBM Corporation, Armonk, NY, USA) was used for statistical analysis with significance level of 0.05. Non-parameter Spearman correlation and Kolmogorov-Smirnov methods were adopted.

### 3. Results and discussion

#### 3.1. Gaseous pollutant and PM emissions

##### 3.1.1. Emission factors

The EFs of  $\text{CO}_2$  and various pollutants measured in this study, including gaseous pollutants ( $\text{CO}$ ,  $\text{CH}_4$ ,  $\text{NO}_x$ , and  $\text{SO}_2$ ), particulate matters ( $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ ), EC, OC, and PAHs from residential HB and CC combustion are listed in Table 1. The calculated MCEs for HB and CC were very close, which were 93.1% and 93.0%, respectively.  $\text{EF}_{\text{CO}_2}$ ,  $\text{EF}_{\text{CH}_4}$  and  $\text{EF}_{\text{NO}_x}$  for HB were significantly lower than that for CC ( $p < 0.05$ ). Although  $\text{EF}_{\text{CO}}$  for HB were lower than CC, the differences were statistically not significant ( $p > 0.05$ ). However, HB had higher  $\text{EF}_{\text{SO}_2}$  values compared with CC ( $p > 0.05$ ), this result could be explained by higher sulfur content in HB (see Table S1) as sulfur content is the main factor affecting  $\text{SO}_2$  emissions during coal burning process (Du et al., 2017a).

The  $\text{EF}_{\text{PM}_{2.5}}$  and  $\text{EF}_{\text{PM}_{10}}$  for CC were about two times higher than that for HB, but the differences of  $\text{EF}_{\text{PM}_{2.5}}$  were not statistically significant between these two fuels ( $p > 0.05$ ). This result was consistent with previous studies, in which  $\text{EF}_{\text{PM}_{2.5}}$  for CC were 1–3 times higher than HB (Chen et al., 2015; Zhi et al., 2008). The ratios of  $\text{PM}_{2.5}$  to  $\text{PM}_{10}$  ( $\text{PM}_{2.5}/\text{PM}_{10}$ ) were  $0.9 \pm 0.4$  for HB and  $0.7 \pm 0.4$  for CC, indicating that high fraction of fine particles could be produced when coal was burned in residential stoves. It should be noted that higher fraction of fine particles was found in HB combustion, which could be partly explained by that the clay content in HB is favorable for fine particulate matter formation (Chen et al., 2015). Analogously, large differences in  $\text{EF}_{\text{EC}}$  and  $\text{EF}_{\text{OC}}$  between HB and CC were observed ( $p < 0.05$ ). Extremely low EFs of EC and OC were found in HB burning, which only accounted for 1.4% and 13% of CC combustion. The ratios of EC to OC (EC/OC, often used for source apportionment) for HB were significantly lower than that for CC, which were  $0.1 \pm 0.1$  and  $0.7 \pm 0.4$ , respectively. The differences of EC/OC were attributed to briquetting technology that could enhance

coal combustion condition and decrease more EC emissions compared with OC (Bond et al., 2002). Chen et al. (2009) previously reported that HB combustion could reduce emissions of EC and OC by a range of 42%–98% and 13%–54%, respectively, compared to CC, thus resulting in lower EC/OC ratios for HB, which was consistent with this study. In addition, calorific value based EFs of pollutants were also calculated based on calorific values reported in the literatures with similar carbon content, results were showed in Table S3. In general, calorific value based pollutant EFs showed similar trends with fuel mass based EFs. However, EFs of  $\text{CH}_4$  for HB combustion was a little higher than that for CC, although not significant ( $p > 0.05$ ).

As mentioned above, the EFs of most pollutants from HB burning were lower than CC, it would be obvious potential for pollutant reductions when switching to HB for residential cooking. In summary, the EFs of  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_4$ ,  $\text{NO}_x$ ,  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ , EC, OC, and PAHs would reduce by a range from 31.7% to 98.6%, but  $\text{SO}_2$  would increase by 12.5%. Coal (HB and CC) is the most widely used energy in Shanxi province due to abundant reserves, emissions from inefficiency coal burning in residential stoves had resulted in severe indoor and ambient air pollution, thus causing adverse outcome on human health. For example, Du et al. (2017b; 2018b) reported that daily personal exposure to  $\text{PM}_{2.5}$  and  $\text{PAH}_{28}$  for residential coal combustion could be as high as  $300 \pm 50 \mu\text{g}/\text{m}^3$  and  $1411 \pm 710 \text{ ng}/\text{m}^3$ , respectively, in rural Shanxi during wintertime. Beside this, due to relatively low combustion efficiency and short of pollutant control devices, pollutant EFs from household stoves could be several orders of magnitude higher than industrial boilers (Zhang et al., 2008). Therefore, the emission reductions from residential coal combustion were crucial. Taking advantage of fuel consumption data from a nationwide energy survey in China (Tao et al., 2018), the total emission mitigation potential by replacing CC with HB for residential burning in Shanxi province in 2012 (the latest available year) was estimated and provided in Table S4. As a result, the reductions of target pollutants ranged from  $6 \times 10^2 \text{ t}$  ( $-1.1 \times 10^4$ – $8.2 \times 10^3 \text{ t}$ ,  $\text{SO}_2$ ) to  $1.5 \times 10^7 \text{ t}$  ( $1.2 \times 10^7$ – $1.9 \times 10^7 \text{ t}$ ,  $\text{CO}_2$ ), respectively. With regard to pollutant emission reduction percentages, the ratios ranged from 2.4% ( $-45.5\%$ – $35.7\%$ ,  $\text{SO}_2$ ) to 98.8% (97.4%–99.5%, EC), indicating an alternative way for pollutant emission reductions through the promotion of HB, consistent with previous studies (Shen, 2015). While for  $\text{SO}_2$ , it is expected that the emission reductions of HB combustion could be achieved by strict desulphurization in briquetting process in the future. Except for pollutant emission reductions, the replacement of CC with HB in residential burning would contribute to the decline of indoor pollutant concentrations. For example, Das et al. (2019a) reported that indoor  $\text{PM}_{2.5}$  concentrations could lower 2–4 times by replacing CC with HB. Therefore, the promotion of HB for household daily usage might be a feasible way to reduce pollutant emissions, improve air quality and decline health risk in rural Shanxi as many households in this area had relatively low income and could not afford clean fuels such as liquid petroleum gas and electricity.

##### 3.1.2. Comparison with previous studies

There are a few studies measured pollutant emissions from residential coal combustion at this stage, among which CO and  $\text{PM}_{2.5}$  were two commonly tested pollutants. However, most previous studies were conducted in controlled condition (laboratory or simulated kitchen) (Chen et al., 2005, 2009; Shen et al., 2010b; Zhi et al., 2008), whereas field measurements were very scarce. It is generally accepted that in

**Table 1**

EFs of  $\text{CO}_2$  and various pollutants measured in this study with arithmetic means and standard deviations.

Fuels	$\text{CO}_2$	CO	$\text{SO}_2$	$\text{CH}_4$	$\text{NO}_x$	$\text{PM}_{2.5}$	$\text{PM}_{10}$	OC	EC	$\text{PAH}_{28}^a$
Unit	g/kg	g/kg	g/kg	g/kg	g/kg	g/kg	g/kg	g/kg	g/kg	mg/kg
HB	$1429 \pm 78$	$84 \pm 35$	$1.8 \pm 1.1$	$9.1 \pm 1.5$	$0.5 \pm 0.3$	$4.5 \pm 1.9$	$6.1 \pm 4.2$	$0.3 \pm 0.4$	$0.02 \pm 0.02$	$29.6 \pm 32.1$
CC	$2558 \pm 62$	$123 \pm 42$	$1.6 \pm 0.2$	$13.2 \pm 1.7$	$1.4 \pm 0.3$	$9.1 \pm 6.5$	$13.4 \pm 7.6$	$2.3 \pm 0.9$	$1.4 \pm 0.1$	$80.6 \pm 68.3$

<sup>a</sup> The sum of  $\text{PAH}_{28}$  in gaseous phase and  $\text{PAH}_{28}$  bounded to  $\text{PM}_{2.5}$ .

laboratory or simulated kitchen studies, researchers followed vigorous procedures to operate stoves following standard testing protocols. While in real-world burning processes, operations are uncontrollable by local residents (Du et al., 2018a).

The  $EF_{CO}$  and  $EF_{PM_{2.5}}$  of coal combustion in cooking stoves from previous literatures were collected (Chen et al., 2006, 2015, 2016; Das et al., 2019a, 2019b; Eilenberg et al., 2018; Du et al., 2017a, 2017b; Li et al., 2016b, 2016c; Liang et al., 2017; Liu et al., 2007, 2016, 2018; Shen et al., 2010b; Tian et al., 2008, 2018; Thompson et al., 2019; Yang et al., 2014; Zhang et al., 2000). The  $EF_{CO}$  for HB and CC ranged from 22.4 to 129 g/kg and 70.9–231 g/kg, with arithmetic means of 78.7 and 125 g/kg, respectively. While for  $PM_{2.5}$  emissions, EFs for HB and CC were in the range of 0.3–5.1 g/kg and 0.3–9.6 g/kg, with arithmetic means of 2.7 and 3.8 g/kg, respectively. The burning of CC yielded higher EFs of CO and  $PM_{2.5}$  compared with HB, which was consistent with this study.

Results from field measurements and controlled condition tests were compared and the results were shown in Fig. 1. As shown in Fig. 1a and b, higher  $EF_{PM_{2.5}}$  and  $EF_{CO}$  were found in field studies, either for HB or CC. As shown in Fig. 1a, the averages  $EF_{CO}$  for CC and HB combustion

from field measurements were 160 and 98.0 g/kg, respectively, around 1.8 and 1.5 times higher than that from controlled condition tests, which were 88.9 and 63.3 g/kg, respectively. Disparities were also found in  $EF_{PM_{2.5}}$  from different measurement conditions, among which field based  $EF_{PM_{2.5}}$  for CC and HB combustion were 5.4 and 3.7 g/kg, respectively, about 2.1 and 2.5 times higher than lab-based results, which were 2.6 and 1.5 g/kg (Fig. 1b), respectively. Similar results were also reported by Du et al. (2018a). EFs played a vital role in emission inventory development, the variations in EFs would lead to large uncertainties in emission inventories, indicating the urgent need for more realistic EFs. This study provides valuable results for the development of emission inventory in region-scale, especially in Shanxi province. However, more field studies should be conducted to measure the EFs of different fuels in different regions in the future for a better inventory calculation. If EFs only measured in well controlled conditions were used to develop emission inventories, there would lead to great underestimation since the lack of EFs data from real world. For example, Zhao et al. (2017) used lab-based EFs to estimate residential  $PM_{2.5}$  emissions in Beijing, the value was much lower than Zhang et al. (2008) measured in field in the same region (1.2 vs 7.4 g/kg). In addition, larger

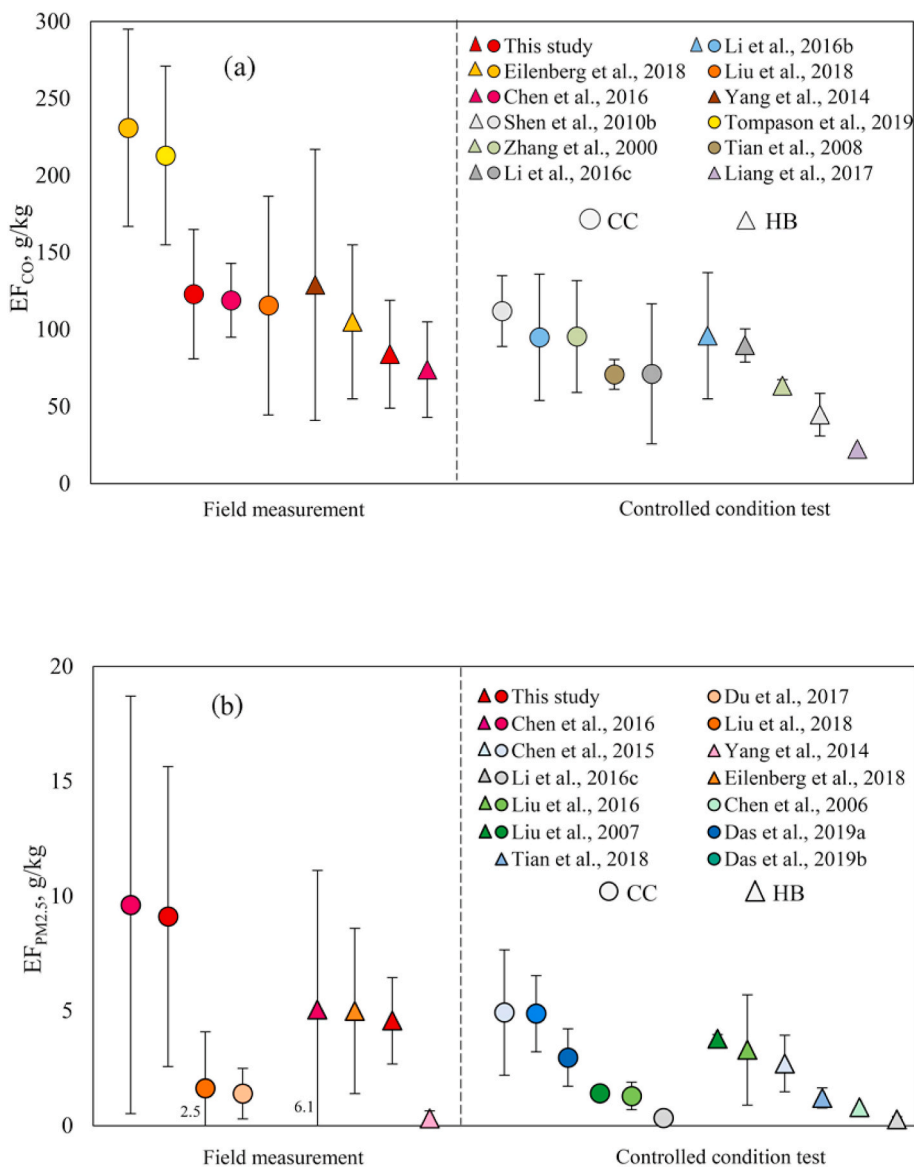


Fig. 1. Emission factors of CO (a) and  $PM_{2.5}$  (b) for HB and CC measured in field and controlled conditions (Data are showed with arithmetic means and standard deviations, the literatures selected for comparison are also shown in the figures).



variations were found in results from field measurements compared with those from controlled condition tests, which was confirmed by coefficient of variations (COVs). For example, the calculated COVs of  $EF_{PM_{2.5}}$  for HB and CC measured in field were 83.3% and 98.5%, respectively, while for those measured under controlled condition, they were only 37.4% and 41.6%. This is partly attributed by the difference of residents' operations (Du et al., 2018a). Large variations were also observed among results from different field tests. In fact, factors affected pollutant emissions in real-world condition are various. For example, the inner structure, size, ventilation, and operational performance varied wildly among different residential stoves, which could affect pollutant emissions (Oanh et al., 2005; Shen et al., 2013a). Besides, in field tests, stoves were operated by local residents, which also varied largely among different residents (Du et al., 2018a). However, real-world emissions are not fully investigated at current stage, which calls for more future studies and large sample sizes to reduce uncertainties of emission inventories.

### 3.2. PAH emissions

#### 3.2.1. PAH emission factors

The EFs of individual PAHs calculated in this study are summarized in Table S5 and the provided results are the sum of gaseous and  $PM_{2.5}$ -bound PAHs. The  $EF_{PAH_{28}}$  (as well as calorific value based EFs) for HB and CC were  $29.6 \pm 32.1$  mg/kg ( $1.6 \pm 1.7$  mg/MJ) and  $80.6 \pm 68.3$  mg/kg ( $2.8 \pm 2.4$  mg/MJ), respectively; with respect to  $EF_{PAH_{16}}$ , the EFs were  $27.0 \pm 28.6$  mg/kg ( $1.5 \pm 1.6$  mg/MJ) and  $62.6 \pm 52.1$  mg/kg ( $2.2 \pm 1.8$  mg/MJ), respectively. PAH emissions from HB burning were much less than that from CC combustion, similar to other target pollutants, which was consistent with previous studies (Chen et al., 2016; Shen et al., 2010a, 2013a, 2013b; Zhang et al., 2008). However, the differences between these two fuels were not significant ( $p > 0.05$ ). Similarly, BaPeq of  $PAH_{28}$  for HB combustion were also lower than that for CC, which were  $12.1 \pm 17.3$  mg/kg and  $20.5 \pm 24.4$  mg/kg. However, the calorific based BaPeq of CC and HB combustion were close, which were  $0.7 \pm 1.0$  mg/MJ and  $0.7 \pm 0.9$  mg/MJ, respectively. It was interesting to note that BaPeq of  $PAH_{16}$  only accounted for  $37.9 \pm 5.2\%$  and  $33.3 \pm 8.4\%$  of the total BaPeq for HB and CC, respectively, even though  $EF_{PAH_{16}}$  accounted for  $94.5 \pm 4.6\%$  and  $83.3 \pm 10.5\%$  of total PAH emissions, respectively, which indicated that non-priority PAHs are more toxic than  $PAH_{16}$ . Therefore, more attention should be paid on non-priority PAHs due to their high toxicity.

Only a few previous studies investigated PAH emissions from residential coal combustion in China, in which most were conducted in controlled conditions. Much higher EFs were again found in field tests when compared with PAH EFs derived from controlled conditions (Table S6). For example, EFs of  $PAH_{16}$  for HB burning measured in field could be 10-fold higher than that from controlled condition tests (Shen et al., 2013a). Larger variations of  $EF_{PAHs}$  could be found in field measurements compared with controlled tests, for example, the averages of COVs for HB and CC measured in field were 120% and 96.9%,

respectively, whereas for EFs measured under controlled conditions, they were only 87.1% and 47.9%, respectively.

#### 3.2.2. Composition profiles and isomer ratios

The percentages of individual compounds to total 28 parent PAHs (composition profile) are presented in Fig. 2. Differences could be observed between the composition profiles of HB and CC. For example, higher RET and CcdP fractions were found for CC combustion, which was consistent with previous emission test (Shen et al., 2015) and exposure air in households burning CC (Du et al., 2020b). PAH emissions from HB were dominated by NAP, FLA and PYR, the sum of these three individuals accounted for  $43.7 \pm 9.8\%$  of total 28 parent PAHs. While for CC, dominant individuals were NAP, PHE and CHR, which accounted for  $50.5 \pm 13.3\%$  of the total. The result of this study was slightly different from Shen et al. (2013a, 2013b), in which dominant PAH compounds for HB and CC burning were PHE, NAP, FLA and NAP, ACY, PHE, respectively. For gaseous PAHs, NAP, ACY, and PHE made the major contribution to these two fuels. While for particle phase PAHs, CHR, PHE, BbF were predominant of HB burning, and FLA, PYR, RET were the dominant species for CC burning. Differences of dominant species in both gaseous and particle phase PAHs could also be found when compared with previous emission studies (Shen et al., 2010a; Wang et al., 2016). The disparities between PAH composition profiles from different tests may be partly explained by fuel properties. It is suggested that differences in coal properties could lead to variations in PAH emission profiles (Zhao et al., 2000). Otherwise, some factors such as combustion efficiency could also affect the composition profiles of PAHs (Wang et al., 2016). The analysis of the PAH composition profiles also found that 2–3 ring PAHs yielded the highest contribution to these two fuels (as shown in Fig. 3), followed by 4 ring and 5–6 ring PAHs, which was consistent with results reported by Liu et al. (2009) and Shen et al. (2013a, 2013b). However, special attention should be paid on 5–6 ring PAHs since these high molecular weight PAHs made the major contribution to total BaPeq because of relatively higher TEF.

The specific PAH isomer ratios are commonly used for source apportionment (Yunker et al., 2002). Six commonly used isomer ratios calculated in this study are listed in Table 2 and significant differences among all these isomer ratios between HB and CC were found ( $p < 0.05$ ). It was suggested that the ratio of FLA/(FLA+PYR) was higher than 0.5 for coal combustion (Yunker et al., 2002). However, contradictory results that FLA/(FLA+PYR) ratios of HB were in the range from 0.48 to 0.51, with mean of  $0.49 \pm 0.01$  were found in this study. Similarly, Shen et al. (2013a, 2013b) and Liu et al. (2018) also reported low FLA/(FLA+PYR) ratios beneath 0.5, which were only 0.42 and 0.47, respectively. As for IcdP/(IcdP+BghiP), the ratio above 0.5, 0.2–0.5 and less than 0.2 indicated coal or biomass burning, traffic and petrogenic source, respectively (Yunker et al., 2002). Fortunately, this was supported by this study. However, contradictory results were reported by Chen et al. (2005) and Zhang et al. (2008), in which IcdP/(IcdP+BghiP) ratios for coal combustion were only 0.33 and 0.35, respectively. Therefore, only using single PAH isomer ratios to distinguish different

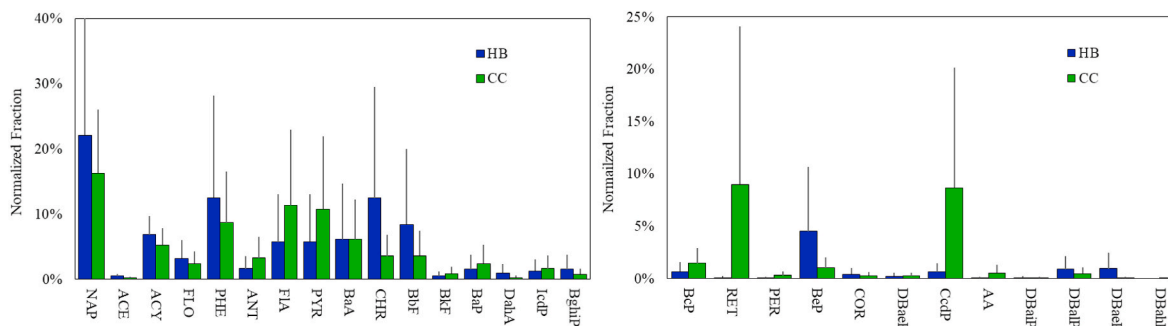


Fig. 2. Composition profiles of PAH emissions from HB and CC combustion.

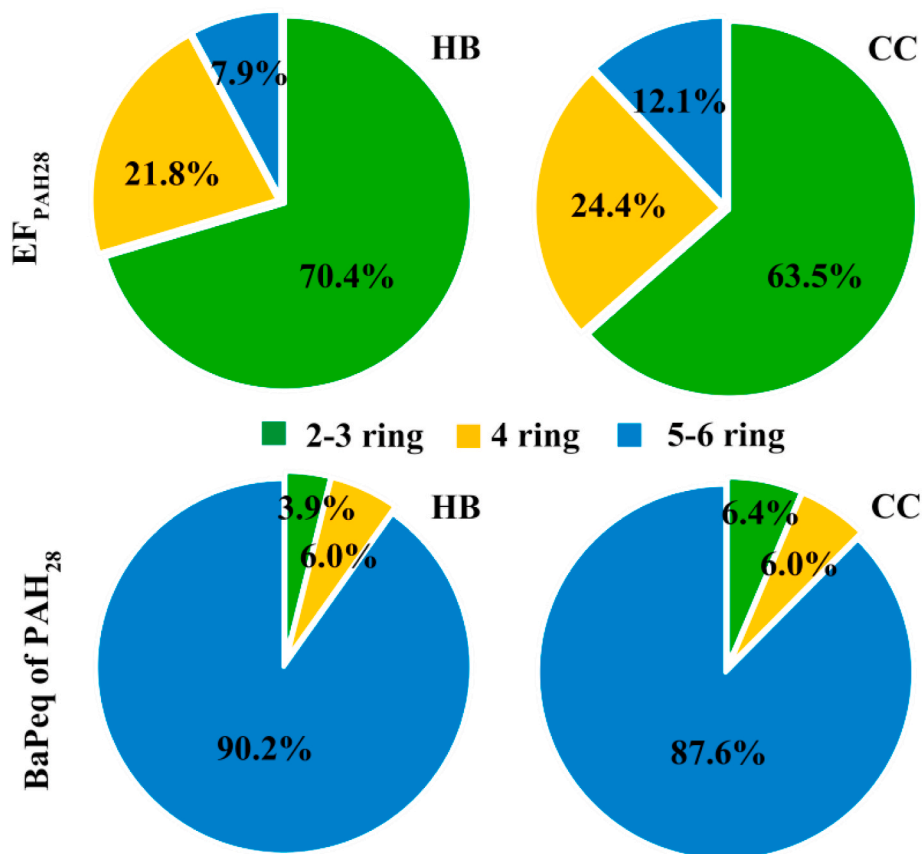


Fig. 3. Contribution of PAHs with different rings to total EFs and BaPeq of PAH<sub>28</sub>.

Table 2

Isomer ratios of PAHs from HB and CC combustion, data shown are arithmetic means and standard deviations.

Isomer ratios	HB	CC
ANT/(ANT+PHE)	0.13 ± 0.03	0.23 ± 0.06
FLA/(FLA+PYR)	0.49 ± 0.01	0.52 ± 0.01
BaA/(BaA+CHR)	0.31 ± 0.02	0.59 ± 0.06
IcdP/(IcdP+BghiP)	0.52 ± 0.07	0.67 ± 0.02
BbF/(BbF+BkF)	0.93 ± 0.01	0.81 ± 0.02
BaP/(BaP+BghiP)	0.51 ± 0.09	0.73 ± 0.05

sources may not be sufficient enough. The most possible reason is the individual PAHs changed differently during the transporting process from source to receptor, then lead to alterations in isomer ratios (Zhang et al., 2005). Besides this, many influence factors such as combustion conditions, coal properties and sampling procedures may also affect PAH isomer ratios (Shen et al., 2013b). Therefore, using single isomer ratios for source apportionment may cause errors and should be used in caution (Krumal et al., 2019). Future experiments are needed to demonstrate the restrictions on application of these PAH isomer ratios for source diagnosis.

### 3.2.3. Gas-particulate partition

EFs of gaseous and particle phase PAH<sub>28</sub> for HB were  $7.6 \pm 4.5$  mg/kg and  $22.0 \pm 25.2$  mg/kg, respectively, while for CC, the EFs of gaseous and particle phase PAH<sub>28</sub> were  $15.6 \pm 8.1$  mg/kg and  $65.1 \pm 54.2$  mg/kg, respectively. Particle phase PAHs were dominant in the PAH emissions for both two measured fuels. Similarly, The BaPeq of particle PAHs made up to 70.0% and 80.6% for the total HB and CC burning, respectively. Gas-particle partition of individual compounds from HB and CC combustion are presented in Fig. S2 and the results were similar between

these two fuels. With the increasing of molecular weight, the contribution of particle phase increased generally. The 2–3 ring PAHs were dominated by gaseous phase PAHs, whereas 4 ring and 5–6 ring PAHs made the major contributions to particle phase PAHs, which was consistent with results from laboratory and field tests (Shen et al., 2010a; Yang et al., 2014). Generally, 2–3 ring PAHs are tented to distribute in gaseous phase (Tobiszewski and Namieśnik, 2012). However, the gas/particle distribution of PAHs could be affected by factors including air temperature (Tasdemir and Esen, 2007). In this study, although the PAHs in particle and gas phase were collected when the PAHs were emitted through the flue gas with high temperature, the samples could be influenced by the ambient temperature ( $\sim 4.3$  °C), which might influence the result of the gas/particle partition. In the future, field studies should be welcomed to achieve a better understanding on the gas/particle partition of PAHs. A study to compare the distribution of PAHs in the chimney (or above) and in the ambient air (in front of the house) at the same time would be interesting.

## 4. Conclusions

High quality pollutant EFs, especially measured in real-world were extremely essential for estimation of pollutant inventories and the climate and health impact caused by solid fuel use. Coal, as a dominant residential energy in China, the EFs based on field measurements were still scarce and overlooked in the past years, thus causing obvious uncertainty in inventory development, climate and health impact estimation. In this study, field measurement was conducted to investigate pollutant emissions from HB and CC burning in residential cooking stoves. The results showed that the burning of HB produced lower pollutant EFs than CC, except for SO<sub>2</sub>. Considering this, the replacement of CC with HB for household burning could be a viable way to achieve pollutant emission reductions in rural households with low income. EFs

from field measurements were usually higher than that from controlled laboratory tests and had higher variations. Therefore, field-based EFs were crucial to compile emission inventories. The fraction of PAH emissions showed a decreasing trend of 2–3 ring, 4 ring and 5–6 ring PAHs for both HB and CC combustion and composition profiles of individual PAHs were different between HB and CC. Particle phase PAHs, which dominated in 4 ring and 5–6 ring PAHs, were prevailed in PAH emissions for HB and CC. The analysis of PAH isomer ratios found that the use of individual isomer ratios as source diagnostic may cause misjudgment and should be used in caution.

#### Credit author statement

Jinze Wang: Methodology, investigation, data curation, writing-original draft preparation, Shanshan Zhang: Methodology, investigation, data curation, writing-original draft preparation, Yan Chen: investigation, data curation and software, Ye Huang: Methodology, investigation, data curation, Zhenglu Wang: data curation and formal analysis, writing-review & editing, Kang Mao: data curation and formal analysis, Weijian Liu: data curation and formal analysis, Wei Du: Conceptualization, supervision, writing-review & editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgments

This work was supported by China Postdoctoral Science Foundation (No. 2019M661425). We sincerely acknowledge the local residents who cooperated with our sampling work. We also acknowledge the editors and anonymous reviewers for their valuable comments.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apr.2020.12.005>.

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