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Effects of biomass briquetting and carbonization on $PM_{2.5}$ emission from residential burning in Guanzhong Plain, China

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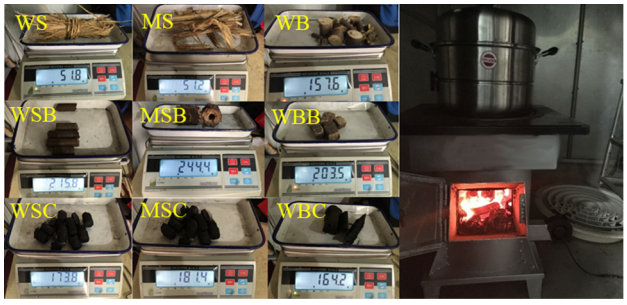
GRAPHICAL ABSTRACT



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ABSTRACT

In this study, maize straw, wheat straw, and wood branches and their processed products (i.e., briquettes and charcoals) were used to investigate the emission factors (EFs) of $PM_{2.5}$ components and the influence of form changing. The EFs of organic carbon (OC) was 241 ± 39.7 to 3920 ± 792 mg kg^{-1}; OC was the largest contributor in $PM_{2.5}$. The EFs of $PM_{2.5}$, OC and elemental carbon (EC) demonstrated that charcoal was the most efficient in reducing $PM_{2.5}$ and carbonaceous fraction emissions, followed by briquettes compared with raw fuels in residential burning use. Among cations, K^+ had the highest EF, whereas Cl^- has the highest EF among anions; the presence of both K^+ and Cl^- was attributable to high abundance in biomass. Cl, K, S, and Na (in descending order) contributed more than 95% of total elements with high EFs; the other elements, particularly heavy metals, had very low EFs. The EFs of total polycyclic aromatic hydrocarbons (PAHs) and saccharides were in the following order of magnitude: raw fuels > briquettes > charcoals. PAHs with four rings (i.e., pyrenes) dominated the total PAHs in $PM_{2.5-}$, whereas among saccharides, levoglucosan had the largest EFs (0.03–110.41 mg kg^{-1}). Briquettes, with simple process and reasonable cost, demonstrated a good ability in decreasing $PM_{2.5}$ emission with potential of 53% (765,000–427,000 t $year^{-1}$), and their emission reduction potential were much higher for straws than for branches. Charcoals demonstrated considerable emission reduction potential for straw (93.7% averagely for $PM_{2.5}$ and compositions) and comparable for wood branches (80% averagely) while its complex process and high cost should be considered. More than 600,000 t of $PM_{2.5}$ and thousands of tons of PAHs and

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saccharides are estimated to be eliminated annually if all raw biomass fuels are processed through carbonization. Considering the efficiency–cost ratios, biomass briquetting may aid clean heating in rural areas, in addition to the "natural gas and electricity replacement strategy," in China and other countries facing similar problems.

1. Introduction

Biomass fuels (including agricultural straws, wood, charcoal, and animal dung) remain commonly used worldwide. More than 3 billion of the poorest people are estimated to rely on biomass fuels burned in inefficient stoves for cooking and heating [1]. In China, more than 288 million t of biomass waste is burned annually; it contains approximately 2.6×10^5-GWh energy, which equals the energy storage of approximately 3.65×10^8 t of coal [2,3]. However, biomass fuel burning, particularly in inefficient conditions, can emit abundant pollutants into the atmosphere including particulate matter (PM) and gaseous pollutants [4–7].

Most particles derived from biomass burning (BB) are PM with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) [8,9]. $PM_{2.5}$ is a detrimental atmospheric pollutant because of its high emission rate and adverse effects on environment and human health [10–12]. Organic carbon (OC), a predominant component of in $PM_{2.5}$ emitted through BB, is derived from organic compounds and has adverse effects on human health and global climate [11,13,14]. Elemental carbon (EC) is a potential toxic components carrier in human and animal respiratory systems [15,16], meanwhile has strong absorption ability to solar radiation and leads to the global warming [17]. BB also produces PM-phase water-soluble inorganic ions (e.g., potassium, sodium, chloride, and calcium) which could act as cloud condensation nuclei (CCN) [18,19]. Polycyclic aromatic hydrocarbons (PAHs), which have constituted a predominant pollutant derived from BB (in > 60% of cases) in China [5,20], has been listed as typical carcinogens by United States Environmental Protection Agency and the World Health Organization. In addition, BB emits numerous levoglucosan-like species, including levoglucosan and mannosan, formed during the pyrolysis of cellulose and commonly used as BB markers in source apportionment studies [21,22].

To further understand BB emission and evaluate the emission reduction effect, reliable emission factors (EFs) must be determined. However, the EFs of $PM_{2.5}$ and particulate-phase pollutant variations depend on many factors, including fuel type and form, water content, and combustion condition [23]. Emission reduction during BB has been widely investigated; however, majority of the related studies have focused on clean stove technology [24–28]. Several studies have reported that fuel processing could also reach the goal of emission reduction from BB. For instance, straw briquetting could enhance fuel density from bulk biomass (10–20 kg m^{-3}) to pellet or bar form (1000–1300 kg m^{-3}), which could slow the burning rate and lower the pollutants formation and emission consequently [7,29]. Biomass charcoal is the product of biomass carbonization left after most of the volatile matter is removed through pyrolysis (~400 °C). It has a high carbon fraction and acts like anthracite in combustion thus has an emission reduction effect [30,31].

Although fuel processing can reduce the EFs of $PM_{2.5}$, systematical studies on emission reduction of different biomass fuel forms are lacking. Meanwhile, because of the diversity of biomass fuels, the same fuel processing technology would lead to different effects on different fuel types. Therefore, in this study, three most popular biomass fuel types in China were selected; each type was obtained three forms: the original form, briquettes and charcoal. Laboratory experiments were then designed for the selected fuels. The study objectives were to evaluate (1) the influence of briquetting and carbonization of typical biomass fuels on the EFs of $PM_{2.5}$ and particulate-phase components emitted from residential cooking/heating and (2) the emission reduction potential of fuel processing technologies to different biomass fuels in residential burning in Guanzhong Plain, China.

2. Methodology

2.1. Fuels and stoves

Three types of typical biomass fuel were selected: maize straw, wheat straw, and wood branches. The original forms of these three fuels were collected in the rural area of Xi'an, China. The briquettes and charcoals of these fuels were processed in Guangzhou Institute of Energy Conversion, Chinese Academy of Science, China. For briquettes, the original bulk biomass was smashed and then pressed at high pressure with moisture of approximately 10% into a bar form. For charcoal formation, the generated briquettes were placed in a furnace for pyrolysis (~400 °C) under isolated oxygen condition. Two-thirds of the mass of the biomass was lost as gas and tar during carbonization; many pollutants, such as $PM_{2.5}$ and PAHs, were also emitted in gaseous, liquid, and particulate phases. The pyrolyzed gas and tar were further used after purification. Because of the low system efficiency and high purification cost, unit cost for charcoal production was about 4 times higher than briquettes which should be considered in overall assessment. The structures of the three fuel types in nine of their forms are shown in Fig. S1; Table 1 lists further information. All collected samples were stored at an ambient temperature (~20 °C) and controlled relative humidity (RH; 35%–40%) prior analysis. Proximate analyses were performed to characterize the samples in their as-received condition. Carbon (C) and nitrogen (N) contents in dry mass, moisture, ash, and volatile matter as well as the fixed C content were determined (Table 1).

The stove used in this study was designed by our research group and equipped with secondary air distribution system (Fig. 1). This clean stove is specially designed for rural residents in Guanzhong Plain; it can fit the main fuel types, including biomass straw, branches, briquettes, anthracite, and charcoals. The stove has a high combustion efficiency for various fuels and can reduce the EFs of $PM_{2.5}$ compared with traditional stoves, as noted in our previous study [32]. The optimal combustion condition can be realized on the basis of differences in the air supply ratios. More detailed information about the stove can be obtained in a 2018 study of Sun et al. [33].

Table 1
Information of collected biomass fuels.

Fuel group	Form (Abbreviation)	Proximate analysis (as received, mass %)			
		Moisture	Ash	Volatile matter	Fixed carbon
Maize straw	Raw (MS)	6.10	4.70	76.00	13.20
	Briquette (MSB)	8.97	6.32	70.00	14.71
	Charcoal (MSC)	6.12	18.89	25.01	49.98
Wheat straw	Raw (WS)	4.39	8.90	67.36	19.32
	Briquette (WSB)	10.28	14.24	65.11	10.37
	Charcoal (WSC)	6.08	25.64	16.95	51.33
Wood branch	Raw (WB)	4.39	2.15	82.96	10.51
	Briquette (WBB)	4.37	2.72	79.66	13.25
	Charcoal (WBC)	3.52	5.59	44.94	45.95

Maize straw and wheat straw group combined to be herbaceous group.

Fig. 1. Fuels and clean stove used in this study.

2.2. $PM_{2.5}$ sampling and chemical analysis

A dilution sampler (Model 18, Baldwin Environmental Inc., Reno, NV, USA) was used to extract the exhaust from combustion chamber. Three nondispersive infrared CO_2 sensors (PP Systems, Amesbury, MA, USA) were used to measure CO_2 concentrations in the undiluted smoke, air used for dilution, and diluted smoke, respectively. The dilution ratios of the dilution system were 0–40 under standard isokinetic sampling. More detailed descriptions of combustion chamber and dilution sampler were provided by Tian et al. in 2015 [34]. $PM_{2.5}$ samples were collected [34] from three parallel channels located downstream of the residence chamber of the dilution sampler, with a flow rate of 5 L min^{-1} $channel^{-1}$. Two 47-mm quartz microfiber filters (Whatman Limited, Maidstone, UK), which were preheated at 900 °C for 3 h before sampling to remove any residual carbon, were used for analyzing OC, EC, water-soluble ions, and organic matter. One 47-mm Teflon-membrane filter (Pall Life Sciences, Ann Arbor, MI, USA), preconditioned at 25 °C ± 0.5 °C temperature and 35% ± 5% RH for 48 h before use, was collected for gravimetric and elemental analyses. The sampled filters were stored in a refrigerator at approximately 4 °C before chemical analysis to minimize the evaporation of volatile components. Before and after sampling, the Teflon-membrane filters were conditioned for 24 h at approximately 25 °C and 35% RH and weighed on a microbalance with a ± 1-μg sensitivity (Sartorius, Göttingen, Germany).

The OC, EC, and their carbon fractions were analyzed following the IMPROVE_A thermal/optical protocol [35]. Water-soluble ions, namely Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}, were detected using a Dionex-600 Ion Chromatograph (Dionex Inc., Sunnyvale, CA, USA). The detection limits of Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-} were 4.6, 4.0, 10.0, 4.0, 5.0, 20.0, 15.0, and 0.5 ppb, respectively. The details of this measurement method were described by Shen et al. [36]. Elemental species were through energy-dispersive X-ray fluorescence (ED-XRF) spectrometry (Epsilon 5 ED-XRF, PANalytical B.V., Netherlands). Detection limits of the 21 elements are listed in Table S1. Details of this procedure have been described elsewhere [37]. PAHs and saccharides were the two main organic matter compounds evaluated; the compounds were analyzed using a gas chromatography/mass spectrometer (Model 7890A/5975C, Agilent Technologies, Santa Clara, CA, USA) simultaneously; this was realized through derivatization reaction in sample pretreatment. Additional detailed information about the pretreatment and settings has been reported by Niu et al. [11]. Overall, 16 preferentially controlled PAHs and 8 saccharides were measured; the detail information is shown in Table S2. For chemical analysis, both quality assurance and control protocol were strictly executed; they included duplicate analysis for 1 of 10 samples, regular calibration, and uncertainty analysis.

2.3. Data analysis

EFs were calculated by dividing the mass of the emissions by the mass of the fuel consumed, and they are expressed in grams of emission per kilogram of consumed dry fuel (g kg^{-1}) [4]. For particulate pollutants, the EFs were calculated as

$$EF_p = \frac{m_{filter}\, V_{total-chimney}}{Q_{filter}\, m_{fuel}}\, DR \quad (1)$$

where EF_p is the EF of particulate pollutant p for the specific fuel type, m_{filter} the mass of pollutant collected on the filter, $V_{total\ -\ chimney}$ the total volume of exhaust flowing through the chimney during the experiment (m^3) at standard temperature and pressure, Q_{filter} the sampling volume through the filter (m^3) at standard temperature and pressure, and m_{fuel} the mass of the burned fuel. The nomenclature of symbols used in equations is listed in Table S3. The dilution rate (DR) of the dilution sampler was calculated based on the CO_2 concentrations:

$$DR = \frac{CO_{2,\ Stk} - CO_{2,Bkg}}{CO_{2,Dil} - CO_{2,Bkg}} \quad (2)$$

where $CO_{2,Stk}$, $CO_{2,Dil}$, and $CO_{2,Bkg}$ represent CO_2 concentrations in stack, diluted smoke, and background air, respectively.

Ion balance calculations can be used for studying the acid–base balance of ions measured in aerosol particle samples. This method was used to evaluate the cation–anion balance of surface-fugitive dust samples. The alkalinity was determined from Eqs. (3) and (4):

$$c\left(cation\ \ microequivalents \frac{1}{m^3}\right) = \frac{Na^+}{23} + \frac{NH_4^+}{18} + \frac{K^+}{39} + \frac{Mg^{2+}}{12} + \frac{Ca^{2+}}{20} \quad (3)$$

$$A\left(\text{anion}\ \ microequivalents \frac{1}{m^3}\right) = \frac{Cl^-}{35.5} + \frac{NO_3^-}{62} + \frac{SO_4^{2-}}{48} \quad (4)$$

2.4. Emission rate estimation

Emission rates were estimated as the burning consumption of the amount of biomass fuels and the corresponding EF. The total consumption of biomass fuels for burning was calculated as

$$M_b = P\ \times = R \times D \times H \times C \quad (5)$$

where M_b is the total mass of biomass used in residential combustion (in

Table 2
Parameters used in the calculation of the total amount of biomass fuels burned in China.

Species	Residue-to-crop ratio[a] (R)	Dry residue fraction[b] (D)	Harvest efficiency[c] (H)	Combustion efficiency[d] (C)
Corn	1.04	0.40	0.92	0.40
Wheat	1.17	0.83	0.86	0.40
Wood branch[e]	1.00	0.90	0.80	0.40

[a] The values were taken from Xie et al. (2011) [70].
[b] The values were cited from Street et al. (2003) [71] and He et al. (2011) [72].
[c] Values refer to Yan et al. (2006) [73].
[d] Refer to Sun et al. (2017) [7].
[e] Calculated as apple tree as representative.

t), P the total production of crop (in t; from China Statistical Yearbook 2016), R the residue-to-crop ratio, D the dry fraction of crop residue, H the harvest fraction of residues by users, and C the ratio of the fuel actually consumed through residential combustion. The parameters are listed in Table 2.

3. Results and discussion

3.1. EFs of $PM_{2.5}$ and carbonaceous fractions

The EFs of $PM_{2.5}$, OC, EC, and CO and the ratios of OC/EC from burning of different fuels are listed in Table 3. The EFs of $PM_{2.5}$ in the herbaceous group were 1.58 ± 0.14 to 9.93 ± 0.34 g kg^{-1}, much lower than those emitted from straw residential smoldering in traditional stove (30.4–46.1 g kg^{-1}) [7]. The wood fuels burning in clean stoves (1.29–1.91 g kg^{-1}) demonstrated much lower $PM_{2.5}$ EFs than did those burning in fireplaces (2.9–9.0 g kg^{-1}) [38]. The EFs of $PM_{2.5}$ were similar to those for improved stoves burning wood and biomass pellets (2.6 ± 2.2 and 2.00–3.66 g kg^{-1}, respectively) [39,40]. These relatively low EFs of $PM_{2.5}$ compared with traditional stoves are attributable to the high combustion efficiency of the clean stove [7,32]. Similar to the results in previous studies, herbaceous fuels produced larger EFs of $PM_{2.5}$ than did wood fuels during combustion ($p < 0.05$) [41]. Meanwhile, for different forms, raw fuels demonstrated the highest EFs, particularly for straws (9.70 ± 0.23 g kg^{-1}), compared with briquettes (4.12 ± 2.22 g kg^{-1}) and charcoals (3.18 ± 1.60 g kg^{-1}). This trend was unclear among the EFs of wood fuels, possibly because the differences between the EFs of raw wood and processed wood fuels were insignificant, even though wood charcoal showed low $PM_{2.5}$ EF (1.29 ± 0.09 g kg^{-1}) [42].

Carbonaceous fractions (i.e., OC and EC) had the most abundant components in combustion emitted $PM_{2.5}$ [43–45]. In this study, the EFs of OC and EC ranged from 241 ± 39.7 to 3920 ± 792 and 95.0 ± 58.7 to 906 ± 86.1 mg kg^{-1}, respectively. These EFs were much lower than those emitted by the residential smoldering of biomass fuels (11.9–17.7 and 0.51–1.1 g kg^{-1} for OC and EC, respectively) [7]. Furthermore, the EFs of OC and EC in the current study were slightly lower than those from biomass open burning (3.3–6.3 and 0.2–0.3 g kg^{-1}, respectively) [6]. A high R^2 of the EFs of $PM_{2.5}$ with those of OC and EC (0.80 and 0.65, respectively) indicated that the low EFs of OC and EC were mainly accompanied by a decrease in $PM_{2.5}$ content, which was influenced by clean stoves (Fig. 2). In herbaceous fuels, similar to the EFs of $PM_{2.5}$, the EFs of both OC and EC were the highest for raw fuels, followed by briquettes and finally charcoals. This is because the high densities of briquettes and charcoals could lower the burning rate and consequently enhance combustion efficiency, which efficiently diminished OC and EC production [28,32]. By contrast, the carbonization process could remove most volatile matter in biomass fuels (Table S1) [30] and could minimize the OC and EC generation during combustion [42].

The OC/EC ratios could reflect the carbonaceous characters in the source-related $PM_{2.5}$ [46–48]. In this study, the OC/EC ratios ranged from 1.86 for wood branch briquette to 4.73 for wheat straw briquettes. These OC/EC ratios were in the range of those reported in relevant literature [41]. Compared with other sources, such as maize straw burned in traditional stoves (10.8–35.1) [7], the OC/EC ratios in this study were much lower ($p < 0.05$), but they were higher than those from coal combustion (1.6–3.0, $p < 0.05$) [44,49] and vehicle emissions (0.5–1.3, $p < 0.05$) [12,50]. Therefore, the OC/EC ratio still has referential meanings in differentiating other sources; however, here, charcoal showed a relatively low value.

The EFs of CO were also calculated (Table 3). The EFs of CO ranged from 12.4 ± 2.3 to 54 ± 1.4 g kg^{-1}, with the highest EFs noted for charcoals, followed by raw fuels and then briquettes. Briquetting caused the biomass fuels to maintain combustion efficiencies higher than those of bulk raw fuels, which led to lower CO emissions. Charcoal burning showed the highest EFs of CO because charcoal has a slow burning rate and high carbon content [51]. The clean stove, with secondary air supply, produced lower CO emissions than did the traditional stoves [52]; however, in this study, the average EFs of CO for the clean stove remained larger than 50 g kg^{-1}, which should not be ignored during the assessment of emission reduction.

3.2. EFs of water-soluble ions

Table 2 listed the EFs of water-soluble inorganic ions from different forms of the selected fuels. The ions occupied 6.6%–13.6% of BB $PM_{2.5}$ mass. In this study, the ion abundance was comparable with that from residential crop residue burning (11%–16%) but significantly lower than that from crop residue open burning (17%–41%) reported previously [7,47,53–55]. On average, the charcoal demonstrated the highest proportion of total ions among the three forms of fuels; however, the difference was statistically insignificant ($p > 0.05$).

The EFs of the ions demonstrated the similar order among the three fuel types and their processed products: K^+ had the largest EFs among

Table 3
Emission factors of $PM_{2.5}$, OC and EC and OC/EC ratios from each fuel burnt.

	$PM_{2.5}$	OC	EC	OC/EC	CO
	g kg^{-1}	mg kg^{-1}	mg kg^{-1}	–	g kg^{-1}
MS	9.47 ± 2.56	3520 ± 1060	906 ± 86.1	3.89	26.6 ± 3.56
MSB	1.90 ± 0.49	669 ± 212	179 ± 67.7	3.75	12.4 ± 2.33
MSC	1.58 ± 0.14	278 ± 2.42	124 ± 8.32	2.24	46.5 ± 9.12
WS	9.93 ± 0.34	3920 ± 792	876 ± 271	4.47	51.0 ± 1.43
WSB	6.35 ± 0.47	726 ± 310	153 ± 28.4	4.73	46.5 ± 2.15
WSC	4.79 ± 0.30	584 ± 32.5	170 ± 8.72	3.43	53.5 ± 13.4
WB	1.59 ± 0.20	638 ± 60.9	291 ± 147	2.19	18.3 ± 2.91
WBB	1.91 ± 0.12	576 ± 5.30	309 ± 107	1.86	15.0 ± 2.61
WBC	1.29 ± 0.09	241 ± 39.7	95.0 ± 58.7	2.54	54.0 ± 1.42

MS: Maize straw; MSB: Maize straw briquette; MSC: Maize straw charcoal; WS: Wheat straw; WSB: Wheat straw briquette; WSC: Wheat straw charcoal; WB: Wood branch; WBB: Wood branch briquette; WBC: Wood branch charcoal.
n = 3 for each fuel burnt.
Up to 3 significant figures were used in EFs.

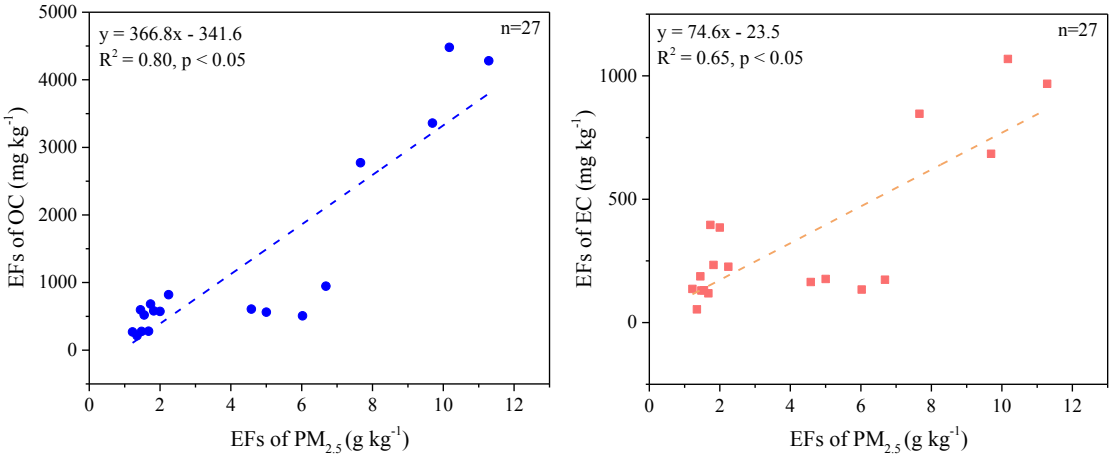

Fig. 2. Correlations between EFs of $PM_{2.5}$ and OC (left) and EC (right).

the water-soluble ions, followed by Cl^- > SO_4^{2-} > NH_4^+ > Na^+ > Ca^{2+} > Mg^{2+} > NO_3^-. According to previous studies, K^+ and Cl^- from BB had high EFs and the emissions and abundances varied with fuel composition and flame temperature [8,42,56,57]. However, the EFs of K^+ and Cl^- were influenced by biomass fuel type and form. For herbaceous fuels, the EFs of both K^+ (59.1–511 mg kg^{-1}; average, 263 mg kg^{-1}) and Cl^- (41.9–454 mg kg^{-1}; average, 197 mg kg^{-1}) are much higher than those from wood fuels (49.6 and 38.9 mg kg^{-1} for K^+ and Cl^- respectively). Meanwhile, the EFs of K^+ and Cl^- showed significant differences among the three fuel forms. In the herbaceous group, the raw fuels (namely WS and MS) yielded the most abundant K^+ and Cl^-, with average EFs of 452 and 359 mg kg^{-1}, respectively. Comparably, the average EFs of K^+ and Cl^- were respectively 137 and 106 mg kg^{-1} in the briquette group and 196 and 121 mg kg^{-1} in the charcoal group—all demonstrating more than 50% decrease in the EFs relative to their raw fuels ($p < 0.05$). The residue ions also followed similar trends: EFs from briquettes and charcoals were lower than those from raw fuels; however, the differences were not as significant as those for K^+ and Cl^-.

As shown in Table 4, the anion/cation ratios In this study ranged from 0.42 to 0.73, consistent with our previous study [7]. The anion/cation ratios in herbaceous group were in the following sequence: raw fuels > briquettes > charcoals. The enhance of alkalinity of $PM_{2.5}$ was possibly due to the steeper decreases in the EFs of anions than in those of cations in processed fuel leading to predominant of cations i.e. K^+, NH_4^+, and even Ca^{2+} [58,59]. The wood fuels here did not show similar trend because of their unusual abundance of NO_3^- and SO_4^{2-}.

3.3. EFs of elements

The EFs of 21 elements were measured (Table 5). All elements occupied 11.1%–37.9% in $PM_{2.5}$ from each fuel; a proportion higher than that of water-soluble ions. The main contributors were Cl, K, S, and Na (in a descending order)—totally contributing to more than 95% of elements. High Cl and K emissions are generally attributable to the high abundance in biomass straws [60,61]; this was because K and Cl are both crucial nutrients for plant growing. The EFs of all other elements were very low level, with those of toxic heavy metals (i.e., Cr and Ni) being even lower, because the straws used were collected from grain crops grown in soils with a generally low heavy metal concentration [62].

For all three different processed fuel types, the total EFs of elements were in the descending order of raw fuels > briquettes > charcoal. In particular, for the trace elements, Cl and K, and some heavy metals, Ba and Pb, a similar sequence was noted, indicating that the processing of biomass fuels could efficiently reduce the EFs of the elements. However, the Σelements/$PM_{2.5}$ ratios demonstrated a reverse sequence. Contributions from the total elements in charcoal-emitted $PM_{2.5}$ were obviously higher than that emitted from raw fuels, as indicated by the higher EF decrease rate for $PM_{2.5}$ compared with that for elements.

3.4. EFs of PAHs and saccharides

The EFs of the 16 PAH species and 8 saccharides from each fuel are listed in Table 6. In total, 15 PAH species were detected based on the absence of naphthalene (NAP). Total EFs of PAHs ranged from 0.17 ± 0.09 mg kg^{-1} in MSC burning to 18.7 ± 5.16 mg kg^{-1} in WS burning. In general, the EFs of total PAHs were in the following descending order: raw fuel > briquettes > charcoals for all three fuel types. Similar results have been reported: charcoal showed much lower PAH abundance in $PM_{2.5}$ compared with wood and briquettes, which could be explained by the high complete combustion led by a low VM content in charcoal [63]. The ratio of EFs between raw fuels and corresponding charcoals can exceed 100, indicating that charcoals

Table 4
Emission factors of water-soluble ions from each fuel burnt (unit of EF: mg kg^{-1}).

	EF_{Na+}	EF_{NH4+}	EF_{K+}	EF_{Mg2+}	EF_{Ca2+}	EF_{Cl-}	EF_{NO3-}	EF_{SO42-}	$\Sigma EF_{ion}/EF_{PM}$	Anion/Cation
MS	22.1 ± 5.26	23.6 ± 1.60	401 ± 6.30	1.88 ± 0.47	21.0 ± 6.10	264 ± 15.9	1.66 ± 0.83	54.2 ± 12.2	8.6	0.65
MSB	10.2 ± 3.37	3.03 ± 1.21	59.1 ± 43.6	0.67 ± 0.15	7.25 ± 1.65	41.9 ± 26.7	0.66 ± 0.24	8.54 ± 2.73	6.6	0.56
MSC	13.3 ± 3.76	2.92 ± 3.45	107 ± 15.9	0.81 ± 0.22	8.94 ± 3.90	69.3 ± 12.6	2.12 ± 0.85	10.3 ± 0.58	13.6	0.57
WS	52.7 ± 12.1	60.2 ± 11.9	511 ± 76.9	1.44 ± 0.47	20.3 ± 7.51	454 ± 19.0	17.9 ± 2.98	72.0 ± 12.0	12.0	0.76
WSB	16.2 ± 2.57	30.7 ± 1.38	214 ± 133	1.00 ± 0.28	15.3 ± 3.94	176 ± 66.3	5.80 ± 4.50	26.3 ± 7.09	7.8	0.67
WSC	16.9 ± 7.97	5.86 ± 0.97	284 ± 8.99	0.82 ± 0.20	7.14 ± 2.02	173 ± 13.8	2.03 ± 0.33	31.9 ± 0.44	10.9	0.66
WB	12.1 ± 3.34	2.05 ± 0.78	45.7 ± 14.1	0.84 ± 0.26	12.5 ± 4.15	24.7 ± 0.34	1.41 ± 0.56	14.3 ± 2.96	7.2	0.42
WBB	13.5 ± 5.00	30.7 ± 3.97	38.7 ± 3.29	0.71 ± 0.34	10.6 ± 3.16	80.2 ± 18.4	10.9 ± 12.9	14.7 ± 3.89	10.4	0.73
WBC	8.41 ± 0.04	1.13 ± 0.56	64.3 ± 18.1	0.46 ± 0.05	4.03 ± 1.41	11.9 ± 5.41	6.09 ± 8.33	30.8 ± 10.4	9.9	0.47

Up to 3 significant figures were used in EFs.

Table 5
Emission factors of elements in $PM_{2.5}$ from each fuel burnt.

mg/kg	MS	MSB	MSC	WS	WSB	WSC	WB	WBB	WBC
Na	17.4 ± 1.62	16.8 ± 6.69	21.8 ± 5.85	76.6 ± 5.09	20.6 ± 13.9	12.7 ± 5.55	9.81 ± 3.57	9.15 ± 5.74	14.6 ± 1.4
Mg	< DL	0.67 ± 0.95	2.46 ± 0.15	3.36 ± 4.75	< DL	< DL	1.75 ± 2.48	0.68 ± 0.96	0.43 ± 0.61
Al	24.7 ± 12.7	6.08 ± 3.12	11.9 ± 1.53	26.1 ± 4.34	14.4 ± 5.07	5.84 ± 8.26	1.72 ± 2.44	3.02 ± 2.21	3.09 ± 0.52
Si	6.77 ± 2.98	7.78 ± 7.02	6.17 ± 4.79	8.45 ± 2.74	4.64 ± 0.48	2.98 ± 1.22	4.21 ± 0.86	3.89 ± 0.48	1.58 ± 0.91
S	67.5 ± 15.7	4.99 ± 1.77	2.64 ± 2.43	46.1 ± 10.8	20.7 ± 8.37	20.5 ± 15.2	14.9 ± 4.40	2.70 ± 2.51	56.9 ± 29.6
Cl	1250 ± 404	333 ± 196	261 ± 110	1630 ± 264	665 ± 343	325 ± 331	149 ± 142	137 ± 152	66.9 ± 7.76
K	1210 ± 414	332 ± 215	243 ± 120	1150 ± 352	496 ± 286	326 ± 296	174 ± 112	45.6 ± 38.3	285 ± 129
Ca	3.78 ± 3.02	1.74 ± 1.35	1.39 ± 1.00	1.36 ± 0.99	4.01 ± 4.43	0.97 ± 0.17	1.65 ± 0.52	0.40 ± 0.21	0.98 ± 0.20
Ti	0.16 ± 0.16	0.08 ± 0.04	0.11 ± 0.03	0.04 ± 0.06	0.07 ± 0.02	0.04 ± 0.03	0.05 ± 0.06	0.03 ± 0.04	0.03 ± 0.01
V	0.03 ± 0.05	< DL	0.02 ± 0.00	< DL	0.01 ± 0.02	< DL	< DL	< DL	< DL
Cr	0.33 ± 0.21	0.15 ± 0.06	0.21 ± 0.05	0.24 ± 0.03	0.15 ± 0.06	0.1 ± 0.02	0.19 ± 0.05	0.15 ± 0.03	0.10 ± 0.03
Mn	0.83 ± 0.78	0.39 ± 0.23	0.42 ± 0.18	0.50 ± 0.33	0.49 ± 0.30	0.23 ± 0.0	0.24 ± 0.04	0.36 ± 0.22	0.21 ± 0.11
Fe	2.42 ± 1.15	1.00 ± 0.19	1.03 ± 0.33	2.13 ± 0.94	1.78 ± 0.92	0.99 ± 0.08	1.32 ± 0.53	1.13 ± 0.25	0.53 ± 0.07
Co	0.15 ± 0.08	< DL	0.06 ± 0.01	< DL	0.01 ± 0.02	0.02 ± 0.01	< DL	0.01 ± 0.01	0.02 ± 0.00
Ni	0.08 ± 0.02	0.03 ± 0.04	0.02 ± 0.00	0.11 ± 0.03	0.01 ± 0.02	0.03 ± 0.01	0.03 ± 0.04	0.04 ± 0.03	0.05 ± 0.01
Cu	0.81 ± 0.31	0.43 ± 0.24	0.44 ± 0.20	1.62 ± 0.35	0.82 ± 0.28	1.19 ± 0.36	0.43 ± 0.04	0.35 ± 0.04	0.27 ± 0.02
Zn	5.87 ± 2.95	9.25 ± 4.94	10.2 ± 3.99	4.19 ± 2.17	2.31 ± 2.21	3.29 ± 0.21	3.12 ± 2.27	1.32 ± 0.65	1.79 ± 0.11
Rb	1.09 ± 0.50	0.66 ± 0.46	1.08 ± 0.25	1.91 ± 0.51	0.94 ± 0.99	1.41 ± 1.57	0.15 ± 0.17	< DL	0.10 ± 0.05
Sb	2.00 ± 0.18	0.52 ± 0.39	1.15 ± 0.15	1.43 ± 0.14	1.35 ± 0.59	0.74 ± 0.47	1.21 ± 0.89	0.69 ± 0.53	0.69 ± 0.11
Ba	5.45 ± 0.85	1.44 ± 0.12	1.28 ± 0.38	3.55 ± 0.63	2.77 ± 0.12	1.29 ± 0.70	2.40 ± 0.11	1.65 ± 0.49	1.55 ± 0.23
Pb	2.66 ± 0.13	2.14 ± 0.63	2.18 ± 0.66	2.72 ± 1.71	1.26 ± 0.62	1.74 ± 1.42	1.69 ± 0.66	4.61 ± 1.07	0.88 ± 0.35
ΣElements/$PM_{2.5}$	27.4%	37.9%	36.1%	19.4%	23.1%	29.7%	23.2%	11.1%	33.9%

Up to 3 significant figures were used in EFs.
< DL denotes below detect limit.

efficiently reduce PAHs emission.

The EFs of individual PAH ranged from 0.01 ± 0.01 to 3.81 ± 1.66 mg kg^{-1}, with a difference more than 300 fold. FLA and PYR were the two PAHs with highest EFs among the measured species, consistent with our previous results [33]. The distribution of PAHs with different ring numbers (Fig. 3) demonstrated that 6-ring PAHs, widely reported as carcinogenic, were mainly detected in $PM_{2.5}$ from raw fuels [11,64]. Thus, the $PM_{2.5}$ emitted from raw fuel burning might have high toxicity to humans. The ring distribution also indicated that PAHs with four rings dominated the total PAHs in all nine groups, even in charcoal, in which the EFs of PAHs were notably low.

The EFs of saccharides were generally higher than those of PAHs, particularly those of levoglucosan (Table 4). The EFs of total saccharides ranged from 0.05 ± 0.07 (WBC) to 135.67 ± 23.86 (MS) mg kg^{-1}; these saccharides were mostly anhydrosugars (i.e., levoglucosan, mannosan, and galactosan). Levoglucosan, the main product of hemicellulose pyrolysis, demonstrated the highest EFs among the eight saccharides measured in the nine fuels (0.03–110.41 mg kg^{-1}). The high EF of levoglucosan could be a useful marker in tracing BB sources [4,65]. Levoglucosan as well as the other saccharides demonstrated EFs in the following order: raw fuels > briquette > charcoal; this was because a majority of hemicelluloses

Table 6
Emission factors of PAHs and saccharides in $PM_{2.5}$ from each fuel burnt.

	MS	MSB	MSC	WS	WSB	WSC	WB	WBB	WBC
NAP	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
ACY	< DL	< DL	< DL	0.04 ± 0.06	< DL	< DL	< DL	< DL	< DL
ACE	< DL	< DL	< DL	3.69 ± 1.79	< DL	< DL	< DL	0.09 ± 0.01	0.05 ± 0.02
PHE	< DL	< DL	< DL	0.02 ± 0.02	< DL	< DL	0.09 ± 0.06	0.01 ± 0.00	< DL
ANT	< DL	< DL	< DL	0.01 ± 0.00	< DL	< DL	0.02 ± 0.01	0.01 ± 0.00	< DL
FLA	0.94 ± 0.67	0.04 ± 0.01	0.03 ± 0.02	1.06 ± 0.11	0.37 ± 0.29	0.11 ± 0.01	0.21 ± 0.06	0.18 ± 0.04	< DL
PYR	1.47 ± 0.91	0.14 ± 0.02	0.15 ± 0.08	3.81 ± 1.66	0.55 ± 0.33	0.25 ± 0.05	0.35 ± 0.07	0.26 ± 0.01	0.11 ± 0.05
BaA	0.63 ± 0.59	< DL	< DL	1.15 ± 0.28	0.17 ± 0.18	0.06 ± 0.01	0.12 ± 0.01	0.04 ± 0.02	0.02 ± 0.00
CHR	0.84 ± 0.75	< DL	< DL	1.51 ± 0.25	0.22 ± 0.23	0.10 ± 0.01	0.16 ± 0.03	0.06 ± 0.02	0.01 ± 0.01
BbF	0.67 ± 0.45	< DL	< DL	1.39 ± 0.17	0.11 ± 0.08	0.06 ± 0.02	0.09 ± 0.00	0.03 ± 0.03	0.09 ± 0.02
BkF	0.71 ± 0.67	< DL	< DL	1.75 ± 0.25	0.12 ± 0.15	0.05 ± 0.01	0.08 ± 0.01	0.06 ± 0.03	0.01 ± 0.02
BeP	0.54 ± 0.43	< DL	< DL	1.02 ± 0.10	0.10 ± 0.08	0.05 ± 0.01	0.07 ± 0.00	0.05 ± 0.01	0.03 ± 0.02
BaP	0.61 ± 0.53	< DL	< DL	1.20 ± 0.24	0.12 ± 0.09	0.06 ± 0.00	0.08 ± 0.00	0.06 ± 0.01	0.03 ± 0.01
IcdP	0.42 ± 0.35	< DL	< DL	1.04 ± 0.10	0.05 ± 0.06	0.02 ± 0.02	0.05 ± 0.01	0.05 ± 0.02	0.02 ± 0.01
DahA	< DL	< DL	< DL	< DL	< DL	< DL	0.06 ± 0.00	< DL	< DL
BghiP	0.50 ± 0.36	< DL	< DL	1.02 ± 0.12	0.06 ± 0.06	0.03 ± 0.00	0.02 ± 0.01	0.03 ± 0.02	0.02 ± 0.01
ΣPAHs	7.32 ± 5.72	0.19 ± 0.04	0.17 ± 0.09	18.7 ± 5.16	1.88 ± 1.54	0.78 ± 0.13	1.39 ± 0.27	0.93 ± 0.23	0.38 ± 0.17
Levoglucosan	110 ± 18.5	0.52 ± 0.14	0.28 ± 0.00	96.4 ± 6.70	5.45 ± 6.25	2.55 ± 1.75	5.46 ± 0.58	0.96 ± 1.36	0.03 ± 0.05
Mannosan	2.10 ± 0.35	0.03 ± 0.01	0.08 ± 0.00	2.12 ± 0.15	0.28 ± 0.33	0.95 ± 0.65	0.36 ± 0.04	0.06 ± 0.08	0.01 ± 0.01
Galactosan	2.99 ± 0.50	0.03 ± 0.01	0.11 ± 0.00	1.97 ± 0.14	0.36 ± 0.41	0.57 ± 0.39	0.29 ± 0.03	0.05 ± 0.06	0.01 ± 0.01
arabitol	18.7 ± 4.20	0.04 ± 0.01	0.03 ± 0.04	0.21 ± 0.05	0.28 ± 0.3	0.12 ± 0.11	0.29 ± 0.06	0.06 ± 0.08	< DL
D-glucose	0.08 ± 0.01	< DL	< DL	0.56 ± 0.24	0.03 ± 0.03	< DL	< DL	< DL	< DL
mannitol	0.44 ± 0.10	< DL	< DL	0.11 ± 0.05	< DL	< DL	0.35 ± 0.11	0.01 ± 0.01	< DL
inositol	0.48 ± 0.07	0.01 ± 0.01	< DL	< DL	< DL	0.02 ± 0.03	< DL	< DL	< DL
sucrose	0.52 ± 0.16	0.13 ± 0.04	0.03 ± 0.01	< DL	0.19 ± 0.11	0.08 ± 0.03	0.13 ± 0.18	0.1 ± 0.14	< DL
ΣSaccharides	135 ± 23.9	0.76 ± 0.21	0.53 ± 0.06	101 ± 7.32	6.58 ± 7.42	4.28 ± 2.96	6.88 ± 0.99	1.23 ± 1.74	0.05 ± 0.07

< DL denotes below detect limit.

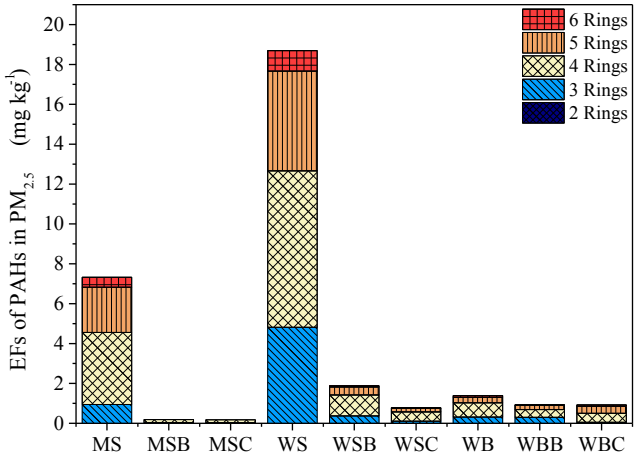

Fig. 3. Ring number distribution of PAHs in different group.

were decomposed during carbonization [30].

Based on the EFs in the present study, the EFs of OC and EC from the boiler of power plant both showed two to four orders of magnitudes lower than those from residential biomass and coal combustion did [66]. It is widely reported that emission control devices (bag-filter precipitator installed at power plant) can efficiently reduce $PM_{2.5}$ emissions with greater than 98% emission reduction efficiency compared with chunk coal residential burning [67,68]. Meanwhile, fuel processing also demonstrated good emission reduction potential in $PM_{2.5}$ and particulate-phase pollutants compared with raw fuels. However, for residential combustion of solid fuels, installing emission control measures in each house was impossible because of economic and social problems. Therefore, fuel processing provided a potential solution for emission reduction in residential solid fuel burning.

3.5. Emission rate estimation and emission reduction evaluation

The emission rates of $PM_{2.5}$ and main particulate-phase components, according to Eq. (3), are listed in Fig. 4. $PM_{2.5}$ emissions were 765,000 t in 2016: 318,000, 427,000, and 20,000 t from maize straw, wheat straw, wood branch burning, respectively. The total $PM_{2.5}$ emission rates estimated were comparable with those reported in Ni et al. (875,000 t $year^{-1}$) [47] but lower than those reported in INTEX-B inventory (4,461,000 t $year^{-1}$) because many other fuels (i.e. coals) were not considered in this study. OC was estimated to have the largest emission rates of 294,000 ton, accounting for 38.4% of total $PM_{2.5}$. The emission rate for EC was lower than that OC with 71,800 t $year^{-1}$ accounting for < 10% in $PM_{2.5}$. Total elements showed very high emission rate (219,000 t $year^{-1}$), which were mostly occupied by K, Cl, S and Na as previously discussed. The sum of the water-soluble ion emission rates was 83,600 t $year^{-1}$, accounting for 11% of the $PM_{2.5}$. These ions can absorb atmospheric moisture and act as cloud condensation nuclei [19,69]. For saccharides and PAHs, the total emission rates were 9767 and 1068 t $year^{-1}$, in which the contribution from herbaceous straws was more than 99%. This was because of the high EFs of PAHs and saccharides in straw burning and large consumption compared with wood fuels.

Considering emission reduction, briquetting efficiently reduced the emission rates of $PM_{2.5}$ and corresponding chemical components. The total $PM_{2.5}$ emission rate was 361,000 t $year^{-1}$, with reduction efficiency of 53% in the assumption of 100% replacement for raw fuels. Comprehensively, the briquettes of maize and wheat straw demonstrated more than 80% emission reduction efficiency for OC and EC compared with raw straw burning and an average of more than 90% for PAHs and saccharides. The emission reduction efficiency for total water-soluble ions and elements was relatively low, but remained at more than 50%, indicating great efficiency. The briquettes of wood branches could not reduce the emission rates of $PM_{2.5}$ and some other components. Briquettes could not reduce the total emission of $PM_{2.5}$; they even increased ion emission more than 100%, although emission reduction efficiency on PAHs and saccharides was good. Charcoals were expected to emit 91,720 t of $PM_{2.5}$ in 2016, with emission reduction efficiencies of 94.4%, 83.9%, and 73.0% for maize straw, wheat straw, and wood branches, respectively. The emission reduction efficiency for PAHs and saccharides could even reach 99% for straws and it exceeded 90% for wood branches. For total ions and elements, the emission reduction performance was relatively low but remained at 90% in the straw group and 60% in the branch group. In summary, biomass fuel processing can efficiently reduce $PM_{2.5}$ and particulate-phase pollution from combustion. Charcoals demonstrated higher emission reduction efficiency than did briquettes because most of the VM were pyrolyzed during carbonization. However, the considerably high cost incurred and pollutant emission during carbonization should be considered when evaluating the emission reduction efficiency and application potential.

4. Conclusions

The EFs of $PM_{2.5}$ and chemical components, namely OC, EC, water-soluble ions, elements, PAHs, and saccharides, from BB in a specially designed clean stove were measured. Three typical biomass fuels, namely maize straw, wheat straw and wood branches, and their processed products, namely briquettes and charcoals, were used to investigate the influence of form changing on the particulate pollutant emission. The EFs of $PM_{2.5}$ ranged from 1.29 ± 0.09 to 9.93 ± 0.34 g kg^{-1}. The EFs of OC and EC both showed a descending order of raw fuels > briquettes > charcoals indicating that fuel processing could reduce the production of $PM_{2.5}$ and carbonaceous fraction during combustion. All water-soluble ions occupied 6.6%–13.6% concentration in $PM_{2.5}$, among which K^+ and Cl^- were the most abundant, as reported previously. All elements were the second largest contributor in $PM_{2.5}$ in this study; Cl, K, S and Na (in a descending order) totally contributed more than 95% of total elements. Among the 15 detected PAHs, the EFs ranged from 0.01 ± 0.01 to 3.81 ± 1.66 mg kg^{-1}, with highest EFs being noted for FLA and PYR. Total EFs of PAHs ranged from 0.17 ± 0.09 (MSC burning) to 18.7 ± 5.16 (WS burning)

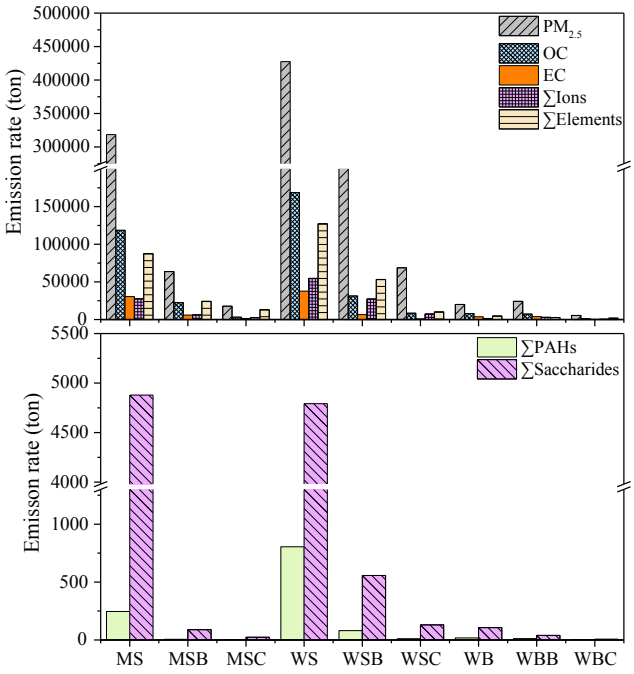

Fig. 4. Emission rate of processed fuels compared to raw fuels (ton).

mg kg^{-1}; the values were in the following descending order: raw fuel > briquettes > charcoals. PAHs with four rings dominated the total PAHs in $PM_{2.5}$ from all nine groups. Levoglucosan showed the highest EFs among the eight saccharides measured in the nine fuels (0.03–110.41 mg kg^{-1}); the EFs were highest in raw fuels and lowest in charcoals. $PM_{2.5}$ emissions were estimated to be 765,000 t in 2016; they included 318,000, 427,000, and 20,000 t from maize straw, wheat straw, and wood branch burning, respectively. When replaced by briquettes, the $PM_{2.5}$ emission rates decreased to 361,000 t $year^{-1}$, with a reduction efficiency of 53%. The briquettes showed better emission reduction efficiency on straw than wood branches because of the high bulk density of straw. Charcoals showed considerably high emission reduction efficiencies for $PM_{2.5}$ (83.8%), OC (93.3%), EC (94.0%), ions (80.1%), elements (79.2%), PAHs (96.2%) and saccharides (97.2%) for both herbaceous and woody fuels. However, considering the high costs and low system efficiency of the carbonization process, briquetting technology would be a proper clean heating method for use in Northern China and other regions facing difficulties in "switching to electricity and natural gas" strategy in rural areas. Furthermore, the effects on health, climate, and environment of $PM_{2.5}$ and other emission components from biomass briquetting and carbonization should be evaluated in the future.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fuel.2019.02.031.

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