Emission factors of organic carbon and elemental carbon for residential coal and biomass fuels in China- A new database for 39 fuel-stove combinations

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ABSTRACT

In recent years many households in northern China's rural areas tend to furnish their houses with water-circulating piping system for heating, which entails mini-boiler stoves to heat water via raw coal chunk or biomass pellets. In this study, consistent efforts were made to obtain first-hand emission factors of organic carbon (EF_{OC}) and elemental carbon (EF_{EC}) for residential solid fuel combustion. A total of 39 fuel/stove combinations, covering seven coals (with different geological maturities), eleven biomass fuels, and five different stoves, were tested. The mean EF_{OC} and EF_{EC} were (4.29 ± 2.33) and (4.43 ± 2.18) g/kg for residential coal combustion, (2.16 ± 4.47) and (0.42 ± 1.01) g/kg for indoor biomass burning. The EFs for tested coal combustion display a "bell shape" with the maximum EF value occurring at bitumite of middle maturity. Coal briquetting in this study led to a significant decrease in EF_{EC} but a notable increase in EF_{OC}, which contradicted with the result from some of previous studies that coal briquetting always leads to relatively low emissions of both OC and EC. The inside reason deserves further clarification. Averaging over the two mini-boiler stoves shows that the introduction of mini-boiler stoves can reduce 5% and 10% of OC from anthracite and bitumite, respectively, and 47% and 53% of EC from anthracite and bitumite, respectively, suggesting that transfer from pure heating stoves to mini-boiler stoves seems unlikely to increase carbonaceous particle emissions, particularly EC. The more significant decline in EF_{EC} than in EF_{OC} indicates that the access to mini-boiler stove for winter heating is very likely to be both a clean air measure and a warming mitigation approach. Updated emission inventories in China for the year of 2014 showed that the OC and EC emissions were 338 Gg and 529 Gg, respectively, from residential coal combustion, and 557 Gg and 79 Gg, respectively, from household biomass burning.

1. Introduction

Coal and biomass fuels are two traditional yet exceedingly important sources of energy. Incomplete burning of these solid fuels releases various pollutants, including particulate matter (PM), organic carbon (OC), elemental carbon (EC, or black carbon, BC) and some toxic organics (e.g., polycyclic aromatic hydrocarbons (PAHs)) (Streets et al., 2003; Bond et al., 2004; Ohara et al., 2007; Zhang et al., 2009; Lei et al., 2011; Wang et al., 2012; Shen et al., 2013a; Zhang et al., 2015a; b), many of which are relevant to human health (Nel, 2005; Zhang and Smith, 2007; Liu et al., 2016), physical education (Calderón-Garcidueñas et al., 2008) and climate changes (Menon et al., 2002; Ramanathan et al., 2007; Ramanathan and Carmichael, 2008; IPCC, 2013; Zhang and Mao, 2015). Inventory studies show that the burning of household solid fuels like coal and biomass contributes a substantial fraction of primary carbonaceous aerosols mainly due to the incomplete

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combustion and poor pollution control measures in household stoves (Bond et al., 2004; Wang et al., 2012; Shen et al., 2013a, 2014; Zhang et al., 2014; Zhi et al., 2015). China is considered the largest contributor of BC emissions in the world and the residential sector in China plays an important role (Lu et al., 2011; Liu et al., 2016).

Laboratory and field measurements in the past decades have shown that emission factors (EFs)of OC and EC for solid fuels burned in China's household sector varied up to several orders of magnitude depending on coal/biomass types, stove styles, burning and sampling conditions, and OC/EC test protocols (Zhang et al., 2000; Zhi et al., 2011; Chen et al., 2005, 2006; 2009, 2015a). Unfortunately actual measurements on such EFs for carbonaceous particles from major emission sources are still very limited, leading to large uncertainties in the estimates of regional and global carbonaceous emissions (Zhang and Smith, 2007; Zhi et al., 2008; Shen et al., 2013a).

Recent rapid progress in economy together with the escalating awareness in air quality and climate brings significant changes in the combustion pattern of household solid fuels in China. The most important change occurs in the pattern of coal burning, such as stove types and coal styles. For example, many households in northern China's rural areas tend to furnish their houses with water-circulating piping system for winter heating, which entails heated water via a chunk-coal burning stove. Such stoves are actually mini-boilers almost purely for room heating purpose unlike traditional simple stoves or structurally improved stoves that are incapable of water heating and circulating (Zhi et al., 2015). The rapid dissemination of those mini-boilers in rural households is inviting an increased raw coal chunk rate and a decreased coal-briquette rate against 5 or 10 years before (Chen et al., 2004, 2005; 2006; Zhi et al., 2008; Shen et al., 2013a; b), which is likely to incur an increase in the emissions of most pollutants, in view of the notion that raw coal chunks release more pollutants than coal briquettes (Chen et al., 2004, 2005, 2009; Zhi et al., 2008, 2009; Shen et al., 2010, 2013a, 2014). Another remarkable change occurs in the burning style of biomass fuels; the shares of compressed biomass fuels (biomass pellets) are on the rise in some areas (Sun et al., 2017) because of strong support from government. This tends to reduce the emissions of major pollutants from household biomass fuels (Shen et al., 2012a; Toscano et al., 2014; Zając et al., 2017; Fachinger et al., 2017). It is thus constructive and meaningful to carry out new measurements to improve the knowledge of emissions from residential solid fuel emissions in China.

The objective of this study is to update the database of EF_{OC} and EF_{EC} based on increasing household mini-boiler stoves for residential solid fuels. Several tens of coal/mini-boiler stove and biomass/stove combinations were arranged for emission measurements to get insight into the new arrays of EFs of OC and EC for new combustion patterns, which is significant to updating the pollutant emissions from residential sector and exploring measures against pollution from household solid fuels.

2. Material and methods

2.1. Fuels and stoves

2.1.1. Coals and stoves
Seven coals covering a wide range of geological maturity were arranged for present study (Table S1 in Supporting Information). Each coal was prepared into two styles: raw-coal chunk and honey-comb briquette. The raw-coal chunks were 3–6 cm in size and the honey-comb briquettes were made by intermixing coal powder with clay (25% of clay) into a 12-hole column, 6 cm in height and 9.5 cm in diameter (Chen et al., 2005, 2015b; Zhi et al., 2008; Sun et al., 2017).

Four household stoves were selected to represent the most popular stove patterns used in China: one of them was specifically for honey-comb briquettes (WJ stove) and the other three were for raw-coal chunks (SC, HD, and LW stoves). Detailed information on these stoves

regarding shape, size, and characteristic structure is given in Supporting Information (Fig. S1) and will be described here briefly. The briquette stove WJ and chunk stove SC are of traditional style widely used especially in past decades in China's households for heating rooms through direct thermal radiation. HD and LW stoves are actually low pressure mini-boilers used for heating rooms by heated water circulating through a piping system. Compared to HD, the LW stove has an additional iron baffle vertically fixed before the flue pipe for lengthening the time of heat exchange between hot flue gas and circulating water (Sun et al., 2017).

2.1.2. Biomass fuels and stove
Eleven kinds of biomass fuels were used in this study. These fuels were classified into 3 groups: 9 crop residues (CR), 1 firewood, and 1 wood-chip pellets (pressed mixture of pine and oak chips) (Table S2 in Supporting Information). Most of them were collected in Shandong province of east China except for rice straw and rape straw that were collected in Heilongjiang and Sichuan provinces, respectively.

Only one biomass-burning stove was selected to represent the most popular stove patterns used for biomass burning in China (Fig. S2 in Supporting Information).

2.2. Fuel combustion and sample collection

The briquettes of 7 coals were only burned in stove WJ and 11 biomass fuels were only burned in biomass-burning stove, whereas all the chunks of 7 coals were burned in the 3 chunk stoves (SC, HD, and LW). The temperature and flow velocity were monitored and recorded during the entire combustion experiments by a thermocouple and a Kurz flowmeter, respectively. The fuel burning processes were managed to simulate the actual habits of rural residents. For example, subsequent to the addition of fuels was a stage of smoldering combustion, followed by a stage of flaming combustion and char combustion (Bond et al., 2007; Zhi et al., 2008, 2009).

Procedures for fuel combustion and emission sampling were similar to that described in our previous study (Sun et al., 2017). Briefly, 2–3 anthracite-briquettes (ca. 600 g each) were ignited in a stove by some solid alcohol gels at first and maintained until combustion turned smokeless. A fuel for test was then added to the stove to begin a sampling cycle. The FPS-4000 system (Fig. S3 in Supporting Information) was applied to collect particles (total suspend particle, TSP) onto quartz-fiber filters (Φ 90 mm, Pallflex, QFF) for further thermal-optical carbon analysis (TOR method, DRI, Model, 2001A). The dilution ratio of flue gas ranged from 30 to 180, depending on the envisaged emission intensity of each combustion process of a fuel as well as on burning conditions. Each sampling period persisted for 100–150 min for coal briquettes, 50–90 min for coal-chunks, and 15–30 min for biomass fuels, respectively. The sampling started when the first batch of a fuel was put into the stove and ended when the fuel was burned out (based on combustion temperatures). The combustion experiment of each fuel was repeated 2–4 times under cold start conditions. Procedure blanks were collected to determine background contamination and were used to correct for all results of this study. All particle-loaded filters were stored in a freezer at −20 °C prior to further analysis.

2.3. Calculation methods

2.3.1. EFs of OC and EC
The mass densities of OC and EC on filters were measured with the TOR method. The values of EFx (g/kg, on dry and ash-free basis) were determined by eq (1) (Chen et al., 2005; Zhi et al., 2008; Sun et al., 2017):

$$EFx = \rho \times A \times 10^{-6}/(M1\text{-}M2) \times F/f \qquad (1)$$

Note, x —OC, EC.

ρ—mass of x per unit area of loaded filter ($\mu g/cm^2$)

A—the area of loaded filter (cm^2)

M1—the mass of a fuel before combustion (kg)

M2—the mass of a fuel after combustion (kg)

F—the total flow rate of flue gas in the chimney (displayed by Kurz)

f—the flow rate of sampled flue gas (determined by FPS-4000)

2.3.2. Calculation of emissions
2.3.2.1. China's residential coal combustion.

$$Ei = \sum_{i=1}^{n} Mi \times EFi \ (\ n = 31) \quad (2)$$

Note, E—the estimated emissions of OC or EC for residential coal combustion.

M—the consumption of residential coal (Gg) (National Bureau of Statistics of China (NBSC, 2015))

EF—the EF_{OC} or EF_{EC} for briquettes burned in WJ stove, or coal chunks burned in SC, HD and LW stoves

i—a specific provincial region (province, autonomous region, or municipality directly under the Central Government)

2.3.2.2. China's household biomass fuel burning.

$$E_j = Q_j \times R_j \times P_j \times EF_j \quad (3)$$

Note, E—the estimated emissions of OC or EC for household biomass burning.

Q—the grain yields (Gg) for main biomass fuels (NBSC, 2015)

R—the ratio of dry residue to production (Lu et al., 2011)

P—proportion of crop residues used as biomass fuels (Tian et al., 2011)

EF—the EF_{OC} or EF_{EC} for biomass fuels. Note: only 7 EFs of crop residues (rice, wheat, corn, bean, cotton, peanut, rape) were used to obtain the EFs for biomass fuels due to the unavailability of grain yields of other 4 biomass fuels in NBSC (2015).

j—a specific provincial region (province, autonomous region, or municipality directly under the Central Government)

3. Results and discussion

3.1. EFs of residential coal combustion

3.1.1. The dominant role of coal ranks in the EFs

All EFs of OC and EC measured in this study using 7 tested coals in 4 stoves are presented in Table 1. Based on Table 1, the effects of coal

rank (e.g., anthracite or bituminite), stove type (popular stove or mini-boiler stove) and coal style (chunk or briquette) can be inferred. The effects of coal rank in the EFs of OC and EC will be elaborated in "3.1.1", and the effects of stove type and processed style will be elaborated latter in "3.1.2" and "3.1.3".

Previous studies have proposed the absolute importance of coal's maturity (rank) on pollutants emissions, particularly regarding OC and EC (Chen et al., 2006, 2009, 2015a; Zhi et al., 2008, 2009). In Table 1, it is very clear that all OC and EC have higher EFs for bituminous coals than for anthracitic coals. This agrees with our previous recognition (Chen et al., 2006, 2009; Zhi et al., 2008, 2009) that coal's geological maturity (represented by V_{daf} value) plays a decisive role in the EFs for residential coal combustion. The relatively low combustion efficiency in household stoves leads to markedly incomplete combustion of volatile matter contained in raw coal, which acts as reactants in producing the final emissions (Zhi et al., 2008, 2009). The dominant role of coal rank on EFs of OC and EC is more directly represented in Fig. 1, where for each stove the mean EF of OC from bituminous coals is about 5–8 times that from anthracitic coals, and the mean EF of EC from bituminous coals is about 7–62 times that from anthracites.

Moreover, a bell-shape relation between V_{daf} and EF is observed for both OC and EC. In Table 1, anthracite coals have been found to have much lower EFs for OC and EC than bituminous coals in general in both briquette and chunk styles, and even more importantly, the EFs do not increase monotonically with V_{daf} but maximize in the middle of the V_{daf} values of tested coals. To be specific, the EF-V_{daf} relation looks like a "bell shape" (Fig. 2). Previous studies proposed a "bell shaped curve" with the maximum EF occurring at a coal of V_{daf} around 30% when coal was burned in household stoves (Zhi et al., 2008, 2009). In this study, the 7 coals, from left to right in Fig. 2a and b, are arranged for increasing V_{daf}. It is obvious that the bell shape profile of EF_{OC} is maintained (Fig. 2a), with EF_{OC} for coal briquettes and chunks (13.63 g/kg and 7.15 g/kg, respectively) both peaking in coal SYS ($V_{daf} = 33.20\%$). Similarly, Fig. 2b shows the bell shape profile of EF_{EC}, with the coal SYS ($V_{daf} = 33.20\%$) again having the highest EF_{EC} values (0.80 g/kg for briquettes and 10.50 g/kg for chunks). It is thus recommendable to minimize the use of middle maturity coal in residential stoves to abate the emissions of pollutants like OC and EC.

Note: From left to right, the values of V_{daf} in the horizontal axis are for NX, CZ, LL, PDS, SYS, XLZ, and LK, respectively. Each EF for briquette is obtained from the sole briquette stove WJ, and each EF for chunk is the average over 3 chunk stoves (SC, HD, and LW).

3.1.2. Influence of stove type on the EFs for residential coal

Stoves used for burning coal are also important element for pollutant emissions (Shen et al., 2013a; Chen et al., 2016; Fachinger et al.,

Table 1
Measured EFs (g/kg) of OC and EC for household coal combustion.

Coal[a]	Briquette in WJ stove		Chunk in SC stove		Chunk in HD stove		Chunk in LW stove		Average over HD and LW stoves (HL)	
	EF_{OC}	EF_{EC}	EF_{OC}	EF_{EC}	EF_{OC}	EF_{EC}	EF_{OC}	EF_{EC}	EF_{OC}	EF_{EC}
Anthracite										
NX	1.24	0.08	0.15	0.03	0.48	0.03	0.42	0.31	0.45	0.17
CZ	1.40	0.07	1.00	0.57	0.31	0.02	0.95	0.28	0.63	0.15
Mean	1.32	0.07	0.57	0.30	0.40	0.03	0.69	0.30	0.54	0.16
sd[b]	0.11	0.01	0.60	0.39	0.11	0.00	0.38	0.02	0.13	0.01
Bituminous										
LL	5.32	0.27	1.05	8.44	1.19	1.73	2.41	2.59	1.80	2.16
PDS	11.49	0.41	8.41	13.09	1.14	0.95	3.35	10.23	2.25	5.59
SYS	13.63	0.80	5.22	11.69	3.80	4.74	12.43	15.05	8.12	9.90
XLZ	12.83	0.71	3.82	13.41	1.13	0.95	4.50	6.03	2.81	3.49
LK	10.41	0.21	1.89	5.26	2.63	0.95	3.97	5.98	3.30	3.46
Mean	10.74	0.48	4.08	10.38	1.98	1.87	5.33	7.98	3.65	4.92
sd[b]	3.27	0.26	2.92	3.47	1.21	1.64	4.04	4.79	2.56	3.04

[a] NX, CZ, LL, PDS, SYS, XLZ, LK were produced in Ningxia, Changzhi, Liulin, Pingdingshan, Shuangyashan, Xinglongzhuang, Longkou, respectively.
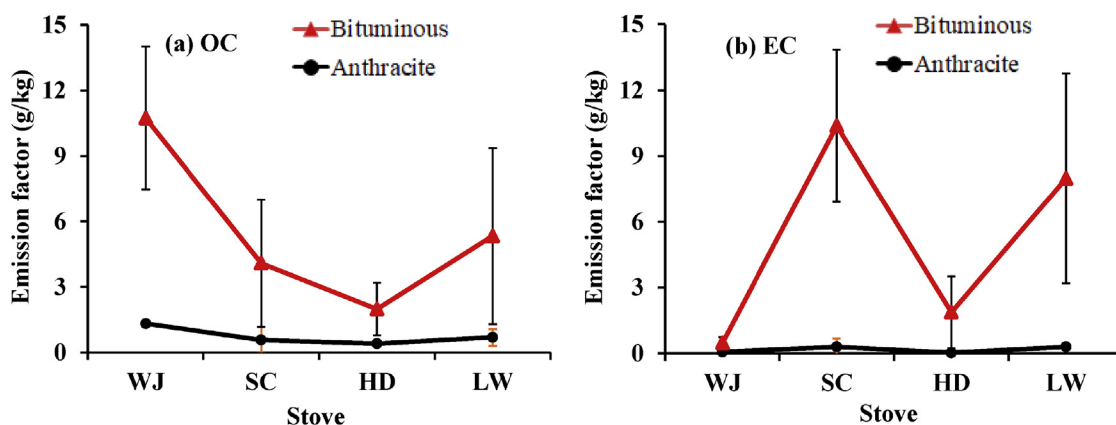
[b] Standard deviation.

Fig. 1. Comparison of EFs between bitumites and anthracites. a) for OC; b) for EC.

2017). In this study, there were in total 3 stoves used for burning chunk coals, among which SC is a simple traditional stove, and HD and LW are 2 mini-boiler stoves. As mentioned in "Introduction", mini-boiler stoves are getting more and more popular in northern China's rural households thanks to their advantage in burning raw coal chunks for rapid household pipe-heating, which in some way complicates OC and EC emission characteristics of household stoves, worthy of a special investigation.

Fig. 3 shows the EFs of OC and EC for the 3 chunk-coal stoves. Data for each stove is the average over two anthracites or 5 bituminous coals (refer to Table 1). In terms of OC, as shown in Fig. 3, LW stove has the highest EF_{OC}, i.e., (0.69 ± 0.38) g/kg for anthracites and (5.33 ± 4.04) g/kg for bitumites, respectively, while HD stove has the lowest EF_{OC}, i.e., (0.40 ± 0.11) g/kg for anthracites and (1.98 ± 1.21) g/kg for bitumites, respectively, indicating that a significant difference exists even between the two mini-boiler stoves and that the popular mini-boiler stoves do not necessarily have higher or lower OC emissions than the traditional ones. In terms of EC, mini-boiler HD releases the least emissions from anthracite (0.03 g/kg) and bitumite ((1.87 ± 1.64) g/kg), while the traditional SC stove releases the most emissions from anthracite ((0.30 ± 0.39) g/kg) and bitumite ((10.38 ± 3.47) g/kg). The HD mini-boiler stove is more effective in abating OC and EC emissions than LW mini-boiler stove, which implies a need to investigate the inner reasons for such result. Averaging over the HD and LW shows that mini-boiler stoves can reduce 5% and 10% of OC from anthracite and bitumite, respectively, and 47% and 53% of EC from anthracite and bitumite, respectively (refer to the last column of Table 1). This may conform to the expectation of the general public that
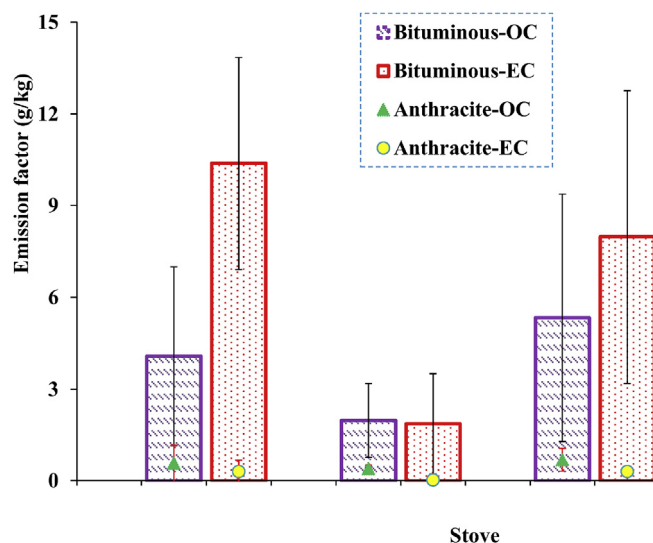

Fig. 3. Comparison of EFs between traditional sample stove and mini-boiler stoves.

transfer from pure heating stoves to mini-boiler stoves seems unlikely to increase carbonaceous particle emissions, particularly EC. In view of the light-absorbing nature of EC (Bond and Bergstrom, 2006; Xu et al., 2009; Chen and Bond, 2010; Streets et al., 2013; Jacobson, 2014; Peng et al., 2016; Hoffer et al., 2017), the more significant decline in EF_{EC} than in EF_{OC} helps identify the access to mini-boiler stove for winter
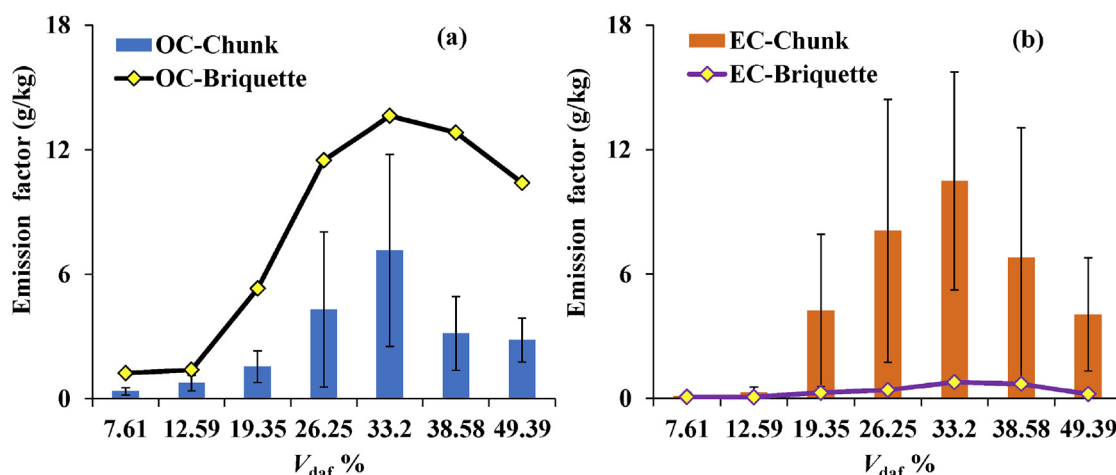

Fig. 2. Variations of EF_{OC} and EF_{EC} for seven coals in two combustion styles. a) OC; b) EC.

heating as both a clean air measure and a warming mitigation approach.

Since each chunk stove (SC, HD, or LW) burned the same suite of coals, the difference in EFs among stoves is only attributable to the difference in individual stoves, especially to stove structure. We noticed that the two mini-boiler stoves are larger in chamber space and longer in chimney than SC stove, which may bring an influence in the combustion efficiency inside stoves. More noteworthy is the iron baffle fixed within the throat of LW stove for higher thermal efficiency, which unexpectedly retarded the smooth flow of flue gas and led to more incomplete combustion in LW stove than in HD stove. This partially explains why the LW stove always had higher OC and EC emissions than HD stove, and reveals an awkward fact: efforts for higher thermal efficiency may reduce combustion efficiency, which consequently brings higher emissions of carbonaceous particles. Stove designers need pay equal attention to thermal efficiency (for energy utilization) and combustion efficiency (for less emission) for win-win result.

3.1.3. Influence of coal briquetting on the EFs of residential coal

Briquetting, an approach to compress powder coal into briquette, has long been considered one of clean coal technologies (CCTs) (Chen et al., 2009, 2015b; Zhi et al., 2009; Li et al., 2016; Zhao and Luo, 2018). Although the effects of briquetting have been investigated in previous studies, finding that coal-briquetting can reduce emissions of BC and some other pollutants (e.g., OC, PM) drastically (Cheng, Y., 1998; Chen et al., 2009, 2015a; Zhi et al., 2009; Shen et al., 2014), new insight and results can still be inferred from this purposeful re-test. Concerning EC, as shown in Fig. 2b, the EF for coal chunk is (5.43 ± 2.67) g/kg, whereas for coal briquette, decreases to (0.40 ± 0.21) g/kg, which indicates a decline of more than 90% and agrees with previous conclusion (Chen et al., 2005, 2009; 2015a; Zhi et al., 2008, 2009; Shen et al., 2013a). It is believed that the briquette's structure (multi-holes with better air-fuel mixing) and composition (including 25% clay) help the complete combustion of coal, leading to less EC released during the burning of briquettes than that of coal chunks (Bond et al., 2004; Zhi et al., 2009).

Contrary to EC that was decreased due to briquetting, OC was considerably increased. The EF_{OC} are (3.15 ± 2.25) g/kg for chunks, but are high as (8.85 ± 2.64) g/kg for briquettes, 2 times up due to briquetting. The difference in the effects of briquetting on OC and EC emissions may result from the different formation mechanisms between OC and EC in coal combustion process. Although such mechanisms have never been specifically addressed, evidences regarding the influence of briquetting in PAHs may indirectly contribute to accounting for the special behavior OC due to briquetting, because PAHs are one important fraction of OC (Cai et al., 2014; Yan et al., 2014; Chen et al., 2015b). After low- and high-ranking coals were burned in a fluidized bed reactor on a laboratory scale, Mastral et al. (1996) believed that the total PAHs emissions were more sensitive to pyrolytic process instead of combustion efficiency. In Chen et al. (2015b), the EFs of 16 parent PAHs, 26 nitrated PAHs, 6 oxygenated PAHs, and 8 alkylated PAHs for coal briquettes were observed to be higher than those for coal chunks, which again strengthened Mastral et al. (1996)'s viewpoint that PAHs are not affected as much by combustion efficiency as EC does, but are more greatly affected by pyrolytic process instead. In fact, briquetting can indeed add to the chances of pyrolytic process because the pulverization increases the surface area of the coal and the compression into a multi-hole cylinder promotes the introduction and circulation of supporting air, which facilitates an easy and close contact of the fuel with the surrounding high temperature air and thereby result in accelerated pyrolysis of the coal organics (Bond et al., 2002; Zhi et al.,2008, 2009). Follow up researches are proposed to focus on the linkage between EF_{PAHs} and EF_{OC} when raw coal chunk is replaced with honeycomb coal-briquette.

The difference in the EFs of OC and EC between this study and previous studies mentioned above may partly result from the different
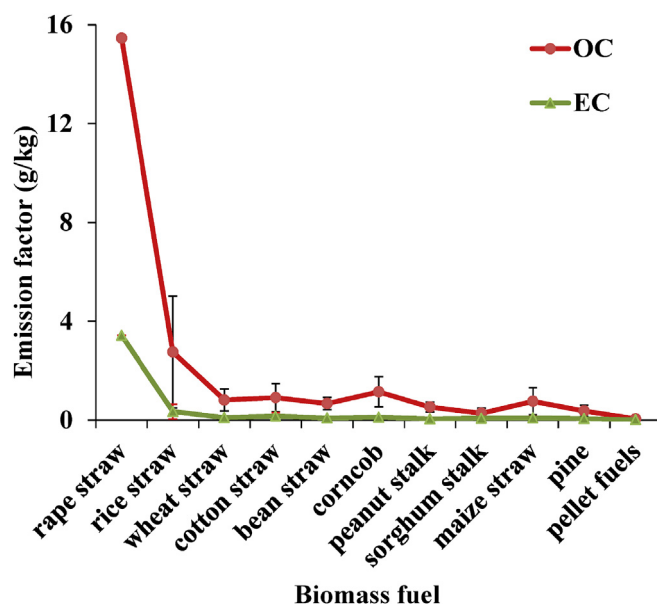

Fig. 4. EF_{OC} and EF_{EC} of household biomass burning in China.

kinds of fuels and stoves for different times. Besides, the difference in analytical protocols may also influence the comparison results (Zhi et al., 2011). EC values analyzed by the IMPROVE protocol are usually higher than those by the NIOSH protocol. This difference is about 2 times for atmospheric aerosols (Chow et al., 2001). Zhi et al. (2011) found that the difference in EC value between IMPROVE and NIOSH protocols was essentially determined by the ratio of EC/TC (TC = OC + EC). Thus, when using the data of EC, we should make clear the measurement method (IMPROVE or NIOSH protocol).

3.2. EFs of household biomass fuels burning

We recognize that the burning condition also affects biomass fuel emissions significantly. In the present study, we tried to simulate the real burning process and condition in households, which covered both flaming and smoldering phases. The EFs reported in this study reflected the whole burning cycle.

The EF_{OC} and EF_{EC} for the 11 biomass fuels used in this study were calculated according to the method described in "2.3" and are presented in Table S3 in Supporting Information. Fig. 4 is plotted based on Table S3. It is very clear that rape straw stands in the first place among the tested biomass fuels (15.46 g/kg of EF_{OC} and 3.43 g/kg of EF_{EC}, respectively), followed by rice straw ((2.76 ± 2.26) g/kg of EF_{OC} and (0.35 ± 0.30) g/kg of EF_{EC}, respectively). The fuel pine, the sole wood fuel in this study, has much lower EF_{OC} and EF_{EC} ((0.37 ± 0.23) g/kg and (0.063 ± 0.055) g/kg, respectively) than CRs. Standing in the last place is the pellet fuel, which has the lowest EF_{OC} and EF_{EC} ((0.050 ± 0.082) g/kg and (0.016 ± 0.027) g/kg, respectively) among all biomass fuels, which agrees with the results of Shen et al. (2012b). Considering that biomass pellet fuels have much lower carbon emissions than raw biomass fuels, which is potential to benefit not only air quality but also climate change, the current promotion of biomass pellets in China should be further encouraged. We also noticed that, in combustion experiments, there were different burning rates among tested household biomass fuels. Herbaceous plants (rape straw, rice straw, wheat straw, cotton straw, bean straw, corncob, peanut stalk, sorghum stalk, maize straw) had larger burning rate than ligneous plants (pine, pellet fuels), coinciding with the order of their EFs mentioned above. In addition because this study does not focus on PAHs emissions, there is no direct evidence here regarding whether biomass pelleting increases PAHs emissions as reported by Perzon (2010) and

Shen et al. (2012a, 2013b). This calls for further investigation.

There are two additional points worthy of noting. The first point relates to the OC/EC ratio. Each EF_{OC} for the 11 individual biomass fuels is higher than EF_{EC}, and the average EF_{OC} and EF_{EC} over the 11 biomass fuels are (2.16 ± 4.47) g/kg and (0.42 ± 1.01) g/kg biomass, respectively, which is consistent with dominant opinion that the ratios of OC/EC for biomass burning are usually higher than those for fossil fuels (Novakov et al., 2005). The second point lies in the relationship between this study and literature. The measured EFs of OC and EC for the wood fuel (pine) are (0.37 ± 0.23) and (0.063 ± 0.055) g/kg, respectively, much lower than those reported by Shen et al. (2012b) for residential wood combustion in a typical cooking stove $((0.60 \pm 0.35)$ of EF_{OC} and (0.94 ± 0.40) g/kg of EF_{EC} for the pine wood combustion). In a field study, Shen et al. (2013a) measured the EFs of OC ((2.2–3.6) g/kg) and EC ((0.91–1.6) g/kg) for the wood (randomly selected wood, without mentioning the wood type) burnt in a simple metal stove in situ of a rural household of rural Shanxi province, which are more than 10 times the values of our study, indicating that biomass fuels combustion in real life may release much more pollutants than laboratory measurement results due to the different kinds of fuel details, stoves, and combustion control manners.

3.3. Comparing EFs of OC and EC between China's residential coal and household biomass fuels

Note: WD-wood (including pine and pellet fuel); AB-anthracite-briquette; AC-anthracite-chunk; CR-crop residue (11 biomass fuels except pine and pellet fuel); BBR-bituminous-briquette; BCH-bituminous-chunk (Based on Table 1 and Table S3 in Supporting Information).

Based on "3.1" and "3.2", Fig. 5 was derived to compare EFs of OC and EC between China's residential coal and household biomass fuels. The EF_{OC} or EF_{EC} for various household coals and biomass fuels used in China exhibits large difference, by and large in the order of WD < AB < AC < CR < BBR < BCH. This implies the similarity to previous studies regarding PAHs and brown carbon (BrC). For example, Chen et al. (2015b) and Sun et al. (2017) found that the emissions of PAHs and BrC for coal briquettes were higher than those for coal chunks. In Shen et al. (2014), the EFs were in the order of anthracite (briquette and chunk) < wood log < brushwood/branches < crop residue < bituminous (briquette and chunk). Yet our EFs of wood are the lowest, which may be attributable to the difference in wood species.
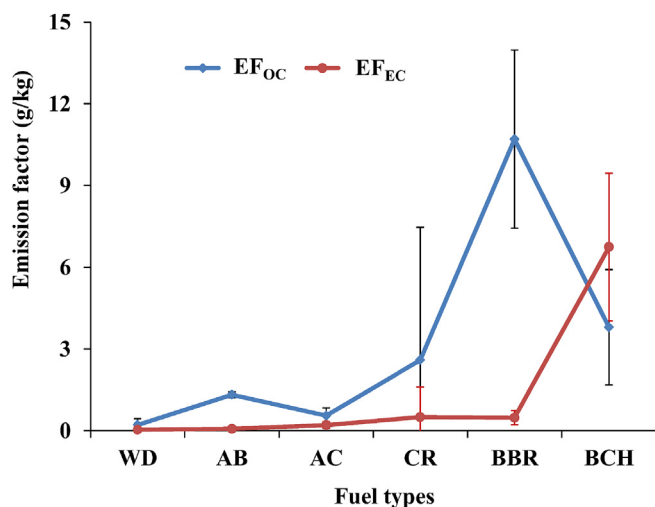
3.4. Emission estimation (E) from China's residential coal combustion and household biomass fuel burning

It is necessary to normalize the EFs for coals or biomass fuels from the measurements in this study to facilitate the calculation of OC and EC emissions from coal or biomass fuel. Coal's EF for OC or EC is merged into a composite value by taking into account the bitumite/anthracite ratio (4:1) and chunk/briquette ratio (4:1) (Chen et al., 2005; Zhi et al., 2008; Sun et al., 2017). Biomass fuel's EF is the simple mean of 11 tested biomass fuels. The composite EFs for OC and EC of coal are (4.29 ± 2.33) g/kg and (4.43 ± 2.18) g/kg, respectively, and the mean EFs for OC and EC of biomass fuels are (2.16 ± 4.47) g/kg and (0.42 ± 1.01) g/kg, respectively. We note that the EFs regarding coal in this study are much higher than those reported by Chen et al. (2015a), who tested five coals (bitumite and anthracite in chunk and briquette styles) burned in three commercial stoves and calculated EF_{OC} and EF_{EC} of (0.90 ± 0.77) g/kg and (1.15 ± 1.19) g/kg, respectively. The EFs regarding biomass fuels in this study are generally lower than those reported (see the description in "3.2"). This suggests the need to track the changes in stoves, coals, and biomass fuels and timely update the EFs of OC and EC to reduce uncertainties of emission inventories and modeling results.

With above EFs and the yearly consumption of coal or biomass fuel in household sector (NBSC, 2015), the emissions of OC and EC from household coal and biomass fuel can be calculated. In 2014, the calculated OC and EC emissions from China's residential coal combustion amounted to 338 Gg and 529 Gg, respectively (Note: EFs for northern China were derived from WJ, SC, HD, and LW stoves and, for southern China, based on WJ and SC stoves). Chen et al. (2006) showed that the calculated results were 478 Gg and 128 Gg for OC and BC (EC) emitted from household coal burning in China during the year 2000, and Zhi et al. (2008) calculated the emissions for OC and EC from household coal combustion in China, 2005, were 357 Gg and 158 Gg, respectively. Obviously, the emissions of OC calculated in this study are comparable to previous ones, but the emissions of EC increased a lot, which seems related to new coal-stove types (including mini-boiler stoves) and increased coal consumption in recent years (Sun et al., 2017; Zhi et al., 2017; Zong et al., 2017). Meanwhile, in the year 2014, the calculated OC and EC emissions from China's household biomass burning amounted to 557 Gg and 79 Gg, respectively, representing an important share in the emission inventories of Tian et al. (2011) (in 2007, the total emissions of BC were 430 Gg for total biomass burning in Chinese mainland) and Lu et al. (2011) (the total emissions of OC and EC from biomass burning in Chinese continental region were 784 Gg and 268 Gg in the year of 2007). We acknowledge that great uncertainties exist in our OC and EC emission inventories due to limited information available. We do need to be careful when using it.

With detailed information on China's household coal/biomass fuel consumption (NBSC, 2015) in 2014 and methodology described in literature (Lu et al., 2011; Tian et al., 2011), the geographical distribution of the emission intensity was obtained (Fig. 6). As shown in Fig. 6a, the top three provinces of EC emission intensity from household coal combustion are Beijing, Tianjin, and Hebei (> 440 kg/km^2), and the last three provinces are Guangxi, Xizang, and Hainan (only 6.01, 0.23, and 0 kg/km^2, respectively). In Fig. 6b, the top three provinces of EC emission intensity from household biomass combustion are Jiangsu, Anhui, and Hubei (> 45 kg/km^2), and the last three provinces are Qinghai, Xinjiang, and Xizang (only 1.24, 0.30, and 0.04 kg/km^2, respectively). The emission intensities of EC from household coal combustion were usually higher for provinces in northeast area than in southwest area, which is related to the fact that there is relatively high yield and consumption of biomass in southern and southwestern provinces.
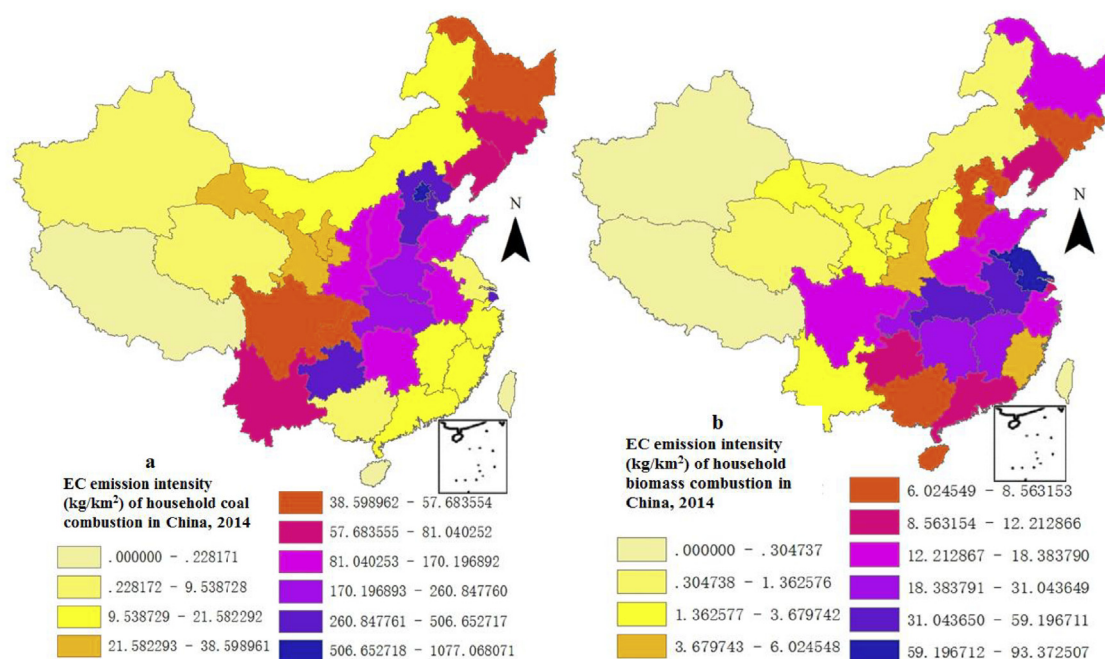

Fig. 5. EF_{OC} and EF_{EC} of different fuel types in China.

Fig. 6. Provincially gridded emission intensity of EC from residential coal and biomass fuels in China's mainland in 2014 (this study). a)coal; b) biomass.

4. Conclusion

Based on fine particle dilution sampling system, EFs of OC and EC were measured for 39 coal/biomass/stove combinations. A new database of EFs for household coal and biomass fuels were acquired, followed by an estimation of the emission inventories of OC and EC. Our measured EFs for OC and EC show that, in the circumstances of this study, coal briquetting decreased EF_{EC} but increased EF_{OC}, which is consistent with the finding on PAHs emissions, but contradicts with conventional notion that coal briquette always has lower emissions of both OC and EC than chunk-coal. Follow up researches are proposed to focus on the linkage between EF_{PAHs} and EF_{OC} when raw coal chunk is replaced with honeycomb coal-briquette and raw-biomass fuel is replaced with biomass pellets so as to clarify whether coal briquetting and biomass pelleting do increase OC and/or PAHs emissions under household burning conditions in China.

Declaration of interest

The authors declare no competing financial interests.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.atmosenv.2018.07.032.

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