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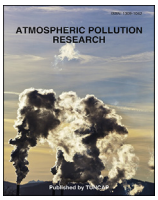

Characteristics of carbonaceous particles from residential coal combustion and agricultural biomass burning in China

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ABSTRACT

Emission factors (EFs) for mass and carbonaceous particles from the residential coal combustion and agricultural biomass burning are measured in the laboratory simulations. Average $PM_{2.5}$, organic carbon (OC) and elemental carbon (EC) EFs from the combustion of a mixture of bituminous and anthracite coals were 6.1 ± 7.1 g kg^{-1}, 1.9 ± 2.3 g kg^{-1} and 2.8 ± 3.8 g kg^{-1}, respectively, and from agricultural biomass burning $PM_{2.5}$, OC and EC EFs were 14.4 ± 3.8 g kg^{-1}, 5.9 ± 2.1 g kg^{-1} and 0.43 ± 0.12 g kg^{-1}, respectively. EFs for the three biomass fuels (wheat straw, maize straw and rice straw) were similar while those from the coals (bituminous coal and anthracite coal) varied with volatile matter content in the fuel. The average OC/EC ratio for agricultural biomass (13.7 ± 2.7) was higher than that for bituminous coal (1.4 ± 1.3) or anthracite coal (6.3 ± 1.3). Carbon fraction profiles showed that EC1-OP was the major product of bituminous coal combustion while OC2 and OC3 were the main emissions from anthracite coal combustion, and OC2, OC3 and EC1-OP were the main products of agricultural biomass burning. $PM_{2.5}$, OC and EC emissions estimates from China in 2012 from coal combustion were 757 Gg, 237 Gg and 343 Gg, respectively, while those from the burning of agricultural biomass were 1238 Gg, 524 Gg and 37 Gg with large differences in per capita emissions among China's provinces.

1. Introduction

Residential coal combustion and biomass burning are important anthropogenic sources for many air pollutants, including primary particulate matters (especially $PM_{2.5}$, particles with aerodynamic diameters ≤ 2.5 μm), organic carbon (OC) and elemental carbon (EC) (Butt et al., 2015; Li et al., 2016; Liu et al., 2015; Streets and Aunan, 2005). With low combustion efficiencies and minimal pollution controls, domestic burning of coal and agricultural biomass for heating and cooking are large contributors to carbonaceous $PM_{2.5}$ in China, especially in rural areas. Take the estimates of Lu et al. (2011) as an instance, the residential sector, including coal and biofuel combustion, accounted for ~69% of the OC and ~51% of the EC in China's primary anthropogenic emissions in 2010.

Better $PM_{2.5}$ emission factors (EFs) for mass, carbon and other components are needed to improve emission inventories and to assist source apportionment (Liu et al., 2015; Watson et al., 2016). There is considerable variability in the published EFs for both residential coal combustion (Table S1, e.g., Chen et al., 2006b, 2015; Shen et al., 2010, 2015; Zhi et al., 2008) and biomass burning (Table S2, e.g. Cao et al., 2008; Li et al., 2009; Shen et al., 2010; Wei

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et al., 2014). These EFs differ owing to the selection of experimental factors, such as fuel types and properties, burning conditions (e.g., smoldering or flaming), stove types (e.g., traditional or improved) and sampling and analysis methods (Bond et al., 2002; Chen et al., 2006a, 2010; Lima et al., 2005; McMeeking et al., 2009; Reid et al., 2005a, 2005b; Ruzer et al., 2012; Watson et al., 2012).

The large variations in the published EFs make it difficult to compile accurate and representative emission inventories. A major problem in this regard is that the EFs used to compile emission inventories often have been based on samples of fuels that are not representative of those currently being used in China, which could create more uncertainty of emission inventory compilation from China (Fu et al., 2013; Ohara et al., 2007). This is especially true for residential burning which is a Chinese-specific large contribution of particulate pollutants. Therefore, up-to-date studies on EFs are needed to refine the emission factor database for China and in so doing develop more robust emission inventories. That information will lead to a more quantitative understanding of the emission characteristics which in turn could be the scientific guidance of taking more targeted and efficient pollution control and management policies.

This study examines laboratory-generated emissions from 2 types of coal and 3 types of agricultural biomass that are used for household cooking and heating in China. $PM_{2.5}$ mass, OC and EC EFs were measured from samples of bituminous and anthracite coals and wheat straw, maize straw and rice straw using typical residential appliance. Study objectives are: (1) to quantify local and regional EFs of $PM_{2.5}$, OC and EC from residential coal and agricultural biomass burning; (2) to characterize carbonaceous fractions of these missions; and (3) to estimate annual $PM_{2.5}$, OC and EC emissions from China's residential sector.

2. Experimental section

2.1. Fuel samples and the test stove

Bituminous and anthracite samples, typical of those in residential use, were collected from major coal producing areas in northern China: (1) Four bituminous coals from Shenmu, Shaanxi Province (B-1), Yulin, Shaanxi Province (B-2), Ordos, Inner Mongolia (B-3), Yinchuan, Ningxia Province (B-4), and (2) two anthracite coals from Wuhai, Inner Mongolia (A-1) and Datong, Shanxi Province (A-2). Biomass from Shannxi and Fujian provinces consisted of: (1) wheat straw (C-1), (2) maize straw (C-2), and (3) rice straw (C-3). Wheat, maize and rice are major agricultural products in China, contributing over 50% of the total agricultural products from 1990 to 2012 (NBS, 2014a).

Fuel samples were stabilized at ~20 °C and 35%–45% RH for 48 h, then ultimate and proximate analyses results according to the national standards of the People's Republic of China, including testing methods for carbon (C), hydrogen (H), oxygen (O), nitrogen (N), sulfur (S), moisture, ash, volatile matter and fixed carbon content, are summarized in Table 1.

A traditional stove was purchased from a local market, and it was specifically selected for the experiments because it is typical of the type most widely used in north China for residential cooking and heating, especially for homes without a central heating system. The stove was 50 cm high, and its hearth had outer diameter of 24 cm, and inner diameter of 12 cm. There was an air-control lip with 6 cm diameter near the bottom, and this was fully opened during the combustion experiments to allow the maximum amount of air to enter the stove during the study (See detail in Fig. S1). This stove was used for both coal and biomass burning.

2.2. Sampling

Experiments were carried out in a laboratory combustion simulator that is described elsewhere (Ni et al., 2015; Tian et al., 2015). The simulator consists of an 8 m^3 chamber instrumented with temperature, pressure and flow velocity sensors coupled to a dilution sampler that permits condensation, growth and aging of emissions. $PM_{2.5}$ concentrations in the chamber were monitored in real time with a DustTrak (Model 8543, TSI Inc, Shoreview, MW, USA) which was normalized to mass using a filter sampler (Wang et al., 2009). Background concentrations in the chamber were <~2 μg m^{-3}, ~0.2%–5% of average $PM_{2.5}$ concentrations during the combustion experiments.

Coal burning EFs were derived from the entire combustion cycle, from ignition to flaming to smoldering and extinction. A honeycomb coal briquette was used to ignite the lumps of raw coal. Honeycomb is a fairly clean type of coal used for both cooking and heating (Zhi et al., 2009). One ignited honeycomb was placed in the stove, the stove was moved into the test chamber when visible smoke disappeared, and background levels were recorded for ten minutes. Pre-weighed samples of the coals (~60–2000 g) were placed on top of the honeycomb, and sampling commenced. For biomass, pre-weighed wheat straw, maize straw and rice straw (~150 g) were inserted into the stove, and ignited with a butane lighter.

After dilution and cooling, $PM_{2.5}$ samples were collected on three parallel channels at 5 L min^{-1} flow rates. Channel 1 contained 47 mm Teflon-membrane filter (2 μm pore size, R2PJ047, Pall Life Sciences, Ann Arbor, MI, USA) while Channels 2 and 3 each contained a pre-baked 47 mm quartz microfiber filters (QM/A, Whatman, Midstone, Kent, England). Three tests were completed for each fuel, yielding a total of 27 experiments. Dilution ratios ranged from 5 to 10 to simulate cooling to near-ambient temperature. Sample durations ranged from 4 to 8 h for coal combustion and ~30 min for biomass burns, sufficient to obtain measureable amounts of $PM_{2.5}$ on the filters.

2.3. Carbon analyses and calculations

Filters were equilibrated at constant temperature (~25 °C) and humidity (35%) and weighed before and after sampling with a high precision electronic balance with a ±1 μg sensitivity (ME 5-F, Sartorius, Göttingen, Germany). Total carbon (TC), organic carbon (OC), elemental carbon (EC) and the thermal carbon fractions (OC1, OC2, OC3, OC4, EC1, EC2, EC3 and OP) were determined on quartz fiber filters using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) following the IMPROVE_A thermal/optical protocol that defines OC = OC1 + OC2 + OC3 + OC4 + OP; EC = EC1 + EC2 + EC3-OP; TC = OC + EC (Chow et al., 2007).

EFs are expressed as the quantity emitted per unit of fuel consumed g kg^{-1} (Delmas et al., 1996). As shown in equation (1), parameters including the dilution ratios (DR), pollutant masses (m_{filter}), sampling durations (t_{sample}), fuel consumption (m_{fuel}), sample volumes (Q_{filter}), stack flow velocities (V_{Stk}) and stack cross section areas (D) were used to calculate the emission factors for the particulate pollutants (EF_p) (See details in Supplemental material S1).

$$EF_p = \frac{m_{filter} \times DR \times t_{sample} \times V_{Stk} \times D}{Q_{filter} \times m_{fuel}} \quad (1)$$

Total China $PM_{2.5}$, OC and EC annual emissions were estimated from the following equation:

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Table 1
Proximate and ultimate analysis of collected fuels (based on air-dry basis, mass %).

Fuel type	Coal						Agricultural biomass		
	Bituminous coal				Anthracite coal		Wheat straw	Maize straw	Rice straw
Fuel identifier	B-1	B-2	B-3	B-4	A-1	A-2	C-1	C-2	C-3
Producing area	Shenmu	Yulin	Ordos	Yinchuan	Wuhai	Datong	Weinan	Weinan	Xiamen
Proximate analysis (mass %, air-dried weight basis)									
Moisture	7.98	6.6	19.47	9.04	4.06	0.88	9.06	8.88	9.02
Ash	7.98	5.09	10.56	4.89	11.06	9.72	7.68	5.53	7.22
Volatile matter	33.2	32.5	24.56	25.56	7.22	6.12	65.99	73.57	66.42
Fixed carbon	50.84	55.81	45.41	60.51	77.66	83.28	17.27	12.02	17.34
Ultimate analysis (mass %, air-dried weight basis)									
Carbon (C)	67.38	71.76	56.61	69.32	76.75	81.37	45.93	47.22	46.12
Hydrogen (H)	3.74	3.88	2.26	3.17	2.28	2.56			
Oxygen (O)	11.8	11.52	9.77	12.74	4.64	3.94			
Nitrogen (N)	0.94	0.87	0.67	0.74	0.86	0.94	0.84	0.65	1.05
Sulfur (S)	0.18	0.28	0.66	0.11	0.35	0.59			

$$E_i = \sum_j M_j \times EF_{i,j} \quad (2)$$

where E_j is the emission a pollutant ($PM_{2.5}$, OC and EC) in Gg; M_j is the quantity of fuel j (bituminous coal, anthracite coal, wheat straw, corn stover and rice straw) burned in Tg; and $EF_{i,j}$ is the emission factor for pollutant i from fuel j in g kg^{-1}.

The total residential coal consumption in China used in our calculations for 2012 was 88 Tg (NBS, 2014b). As fractions of bituminous and anthracite coals in the total coal burned are unavailable, their ratios in total production of raw coal (roughly 78.1% bituminous and 17.6% anthracite) were used to estimate their usage in the residential sector (Chen et al., 2009). The quantity of agricultural biomass burned for domestic purposes was estimated by multiplying the total mass of agricultural biomass by the proportion of agricultural biomass burned as fuel. The total production of agricultural biomass can be calculated from the crop yields (NBS, 2014a), the residue-to-crop ratio and the dry fraction of crop residue. The proportion of agricultural biomass burned as fuel was taken from a survey by Gao et al. (2002). The quantities of coal and agricultural biomass burned for domestic heating and cooking for each province in China in 2012 are presented in Tables S3 and S4.

3. Results and discussion

3.1. Emission factors for carbonaceous particles

$PM_{2.5}$, OC and EC EFs from these experiments are presented in Table 2. Bituminous coals EFs were 10.9 ± 8.1 g kg^{-1}, 3.4 ± 2.6 g kg^{-1} and 5.0 ± 4.7 g kg^{-1} for $PM_{2.5}$, OC and EC, respectively, which are 30–250 times higher than the corresponding EFs for anthracite coals ($PM_{2.5}$ = 0.41 ± 0.08 g kg^{-1}, OC = 0.11 ± 0.02 g kg^{-1} and EC = 0.02 ± 0.00 g kg^{-1}). These differences can be attributed the larger fraction of volatile matter content in the bituminous coals (Table 1). Statistically, significant ($p < 0.01$) positive correlations were found between the volatile matter content and the EFs for $PM_{2.5}$ ($R^2 = 0.92$), OC ($R^2 = 0.78$) and EC ($R^2 = 0.94$). Shen et al. (2010) also found that the coal volatile content of coal explained 48% of emission factor variances.

Bituminous coal EFs and volatile matter contents were more variable than those for the anthracite coals. B-1 with a high volatile matter content (33%) emitted seven times that of sample B-4 with a medium volatile content (26%). Emission inventories might be improved by taking this relationship taken into account.

Table S1 shows there were a wide range of EFs (4.94 g kg^{-1} for $PM_{2.5}$, 0.59–9.78 g kg^{-1} for OC and 0.38–9.81 g kg^{-1} for EC) for bituminous coals, and a smaller range (1.04 g kg^{-1} for $PM_{2.5}$, 0.03–0.28 g kg^{-1} for OC and 0.01–0.02 g kg^{-1} for EC) for anthracite coals (Chen et al., 2006b, 2015; Shen et al., 2010, 2015; Zhi et al., 2008). EFs from this study fall within these ranges. Bituminous coal OC and EC EFs from this study are lower than those reported by Chen et al. (2006b) and Zhi et al. (2008), but higher than those of Chen et al. (2015) and Shen et al. (2010) (Table 2, Table S1). Fig. S2 compares EFs and volatile matter content from several studies. The relationship between EFs and volatile matter content is not as consistent as it is in this study. This reflects the large range in experimental conditions, such as appliance efficiency, oxygen supply conditions and measurement method (Bond et al., 2002; Chen et al., 2006b; Zhi et al., 2009).

Agricultural biomass burning EFs in Table 2, for $PM_{2.5}$, OC and EC in order were 11.0 ± 0.8 g kg^{-1}, 3.7 ± 0.3 g kg^{-1} and 0.29 ± 0.02 g kg^{-1} for wheat straw, 15.7 ± 4.4 g kg^{-1}, 6.4 ± 1.8 g kg^{-1} and 0.48 ± 0.07 g kg^{-1} for maize straw and 16.5 ± 3.2 g kg^{-1}, 7.7 ± 1.3 g kg^{-1} and 0.52 ± 0.06 g kg^{-1} for rice straw. There were no significant differences ($p > 0.05$) in $PM_{2.5}$, OC and EC EFs among the three types of agricultural biomass. $PM_{2.5}$, OC and EC EFs averaged for equal amounts of the three types of biomass were 14.4 ± 3.8 g kg^{-1}, 5.9 ± 2.1 g kg^{-1} and 0.43 ± 0.12 g kg^{-1}, respectively. OC and EC emissions are comparable to published values (Table S2), which range from 0.85 to 6.87 g kg^{-1} for OC and 0.09–2.46 g kg^{-1} for EC (Cao et al., 2008; Habib et al., 2008; Li et al., 2009; Saud et al., 2012; Shen et al., 2010; Wei et al., 2014).

As note, moisture, size, bulk densities of agricultural biomass and combustion temperature can affect $PM_{2.5}$, OC and EC emissions

Table 2
$PM_{2.5}$, OC and EC emission factors from residential coal combustion and agricultural biomass burning (Averages and standard deviations of 3 tests in g kg^{-1}).

Combustion type	Fuel ID[a]	$PM_{2.5}$	OC	EC
Bituminous coals[b]	B-1	18.5 ± 2.7	4.6 ± 0.7	10.6 ± 2.4
	B-2	17.1 ± 2.4	6.3 ± 2.0	6.8 ± 0.6
	B-3	3.5 ± 0.3	1.7 ± 0.4	0.63 ± 0.33
	B-4	2.2 ± 0.4	0.58 ± 0.07	0.45 ± 0.17
	Average	10.9 ± 8.1	3.4 ± 2.6	5.0 ± 4.7
Anthracite coals[b]	A-1	0.46 ± 0.09	0.10 ± 0.02	0.02 ± 0.00
	A-2	0.36 ± 0.03	0.11 ± 0.02	0.02 ± 0.00
	Average	0.41 ± 0.08	0.11 ± 0.02	0.02 ± 0.00
Agricultural biomass[c]	C-1	11.0 ± 0.8	3.7 ± 0.3	0.29 ± 0.02
	C-2	15.7 ± 4.4	6.4 ± 1.8	0.48 ± 0.07
	C-3	16.5 ± 3.2	7.7 ± 1.3	0.52 ± 0.06
	Average	14.4 ± 3.8	5.9 ± 2.1	0.43 ± 0.12

[a] See Table 1 for fuel identifiers.
[b] Emission factors calculated on a dry-weight, ash-free basis.
[c] Emission factors calculated on an air-dried basis.

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and lead to differences between studies (Dhammapala et al., 2006, 2007; Li et al., 2007; McMeeking et al., 2009). EC emission factors in this study are lower than the values reported by Shen et al. (2010), which is possibly due to low fuel moisture and the restricted air supply in their study. Lower moisture content favors higher combustion temperatures, and a restrained air supply can cause low combustion efficiency. Both of them are favorable to EC formation leading to high EFs.

Based on limited data, Andreae and Merlet (2001) reported EFs for $PM_{2.5}$ (7.2 ± 2.3 g kg^{-1}), OC (4.0 ± 1.2 g kg^{-1}) and EC (0.59 ± 0.37 g kg^{-1}) for biofuels (including agricultural biomass). Streets et al. (2003) assumed OC and EC EFs of 5 g kg^{-1} and 1 g kg^{-1}, respectively, for residential biofuels and 0.12–3.00 g kg^{-1}, and 0.12–3.7 g kg^{-1} for residential coals. These values have been widely used for emission inventories, while recognizing their limitations (Ohara et al., 2007; Yan et al., 2006). $PM_{2.5}$ and OC EFs for agricultural biomass reported here are higher than those of Andreae and Merlet (2001), but EC EFs are lower. Coal OC, and EC EFs in this study are within the ranges reported by Streets et al. (2003). Shen et al. (2014) compiled and compared $PM_{2.5}$, OC and EC EFs for solid fuels burned in China. Compared with the classified group (including anthracite lumps, bituminous lumps and crop residue) reported in Shen et al. (2014), this study's EFs for $PM_{2.5}$, OC and EC from agricultural biomass burning were close to the upper limit, and comparable with EFs for coal burning.

3.2. Primary OC/EC ratios and carbon fractions

OC/EC ratios from these tests are compared in Table 3 to evaluate their utility in source profiles (Cao et al., 2005, 2007; Chow et al., 2004; Novakov et al., 2000; Ram et al., 2008; Saarikoski et al., 2008). OC/EC enrichments have been used to separate primary organic carbon (POC) from secondary organic carbon (SOC) (Downward, 2015). This source apportionment is becoming less certain owing to the wide range of OC/EC ratios found in this and other studies (Chow et al., 2010).

The average OC/EC ratio from bituminous coal combustion (1.4 ± 1.3) was significantly lower than that from anthracite coals (6.3 ± 1.3) ($p < 0.001$). These ratios fall within the range reported for similar fuels in household combustion, that is, 0.32–13.6 for bituminous coal and 1.49–14.08 for anthracite coal (See Table 3, e.g., Chen et al., 2006b, 2015; Zhang et al., 2012; Zhi et al., 2008). OC/EC ratios from bituminous coals were lower than those from anthracite coals (Table 3), which has implications for estimating the radiative forcing from the coal combustion (Jacobson, 2001).

For agricultural biomass, the OC/EC ratios ranged from 13 to 15, and these ratios were higher than those measured for coal combustion in our study, which were comparable with previous findings (Bond et al., 2012; Hansen et al., 2005). Comparisons with other studies show consistency with the results reported for agricultural biomass burned in residential stoves by Li et al. (2009), but our values were little higher than those reported by Cao et al. (2008) (Table 3). Li et al. (2009) used improved biofuel stoves, and their results showed that the flaming phase dominated with high-temperature producing more EC and lower OC/EC ratios. Cao et al. (2008) burned piles of crop residue with a lower O_2 supply, resulting in incomplete combustion, higher EC and lower OC/EC ratios. Open burning for land clearing and biomass disposal, emissions showed greater variability for OC/EC ratios. Li et al. (2007) reported a low ratio of five for the open burning of wheat straw during a laboratory measurement, as compared to 11.2 for maize straw. Hays et al. (2005) obtained OC/EC ratios of 2.4 for wheat straw and 52.6 for rice straw for open burning. Different protocols also affect OC/EC ratios (Chow et al., 2001, 2007; Watson et al., 2005).

The thermal carbon fractions (OC1, OC2, OC3, OC4, EC1, EC2, EC3, and OP) yielded by the IMPROVE_A protocol are useful parts of the source fingerprint (Cao et al., 2005; Chow et al., 1993, 2004; Kim et al., 2003a, 2003b). And their abundances in these tests are compared in Fig. 1. The quartz filters from bituminous coal (B-1, B-2, B-3 and B-4) combustion were highly loaded with EC1-OP, which accounted for 24–69% of the TC whereas the anthracite coal (A-1, A-2, and A-3) filters were highly loaded with OC2 and OC3, which together made up over 50% of the TC. Several fractions (OC2, OC3 and EC1-OP) dominated the biomass samples, EC2 (i.e., the 740 °C fraction) has been found to dominate the EC emissions from wood burning (Chen et al., 2007). A small fraction of the OC1 may be lost due to evaporation of this fraction during aging (Chuang et al., 2013). Bituminous coal and agricultural biomass samples had significant amounts of OP, which may relate to the polar OC, which is common in biomass burning emissions (Yang and Yu, 2002).

Table 3
Comparison of OC/EC ratios from residential coal combustion and agricultural biomass burning.

Fuel type	This study	Literature
Coal		
Bituminous coal	1.4 ± 1.3*	1.14–13.6[a,***], 0.38–0.44[b,**], 0.32–5.56[c,***]
Anthracite coal	6.3 ± 1.3*	7.78–14.08[a,***], 1.49[b,**], 3.7[c,***], 7.6[d,**]
Agricultural biomass		
Wheat straw	12.9 ± 2.1*	6.3–11.1[e,**], 8.2[g,**]
Maize straw	13.2 ± 2.9*	4-10[e,**], 2.4[g,**]
Rice straw	15.0 ± 3.6*	10-12.5[e,**], 4.1[g,**]

Data from: a. Chen et al., 2006b; b. Chen et al., 2015; c. Zhi et al., 2008; d. Zhang et al., 2012; e. Li et al., 2009; g. Cao et al., 2008; The asterisks (*) indicate the different protocols used in carbon particles analysis: * represents IMPROVE_A; ** represents IMPROVE, and *** represents NIOSH.

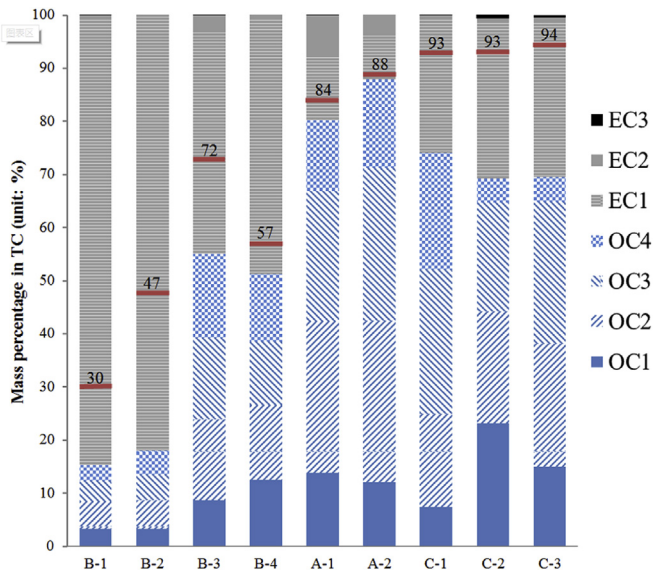

Fig. 1. Mass percentage of thermal carbon fractions in TC on the collected filters from residential coal combustion and agriculture biomass burning following IMOPROVE_A protocol (Chow et al., 2007). OC1 to OC4 are OC evolved in 100% helium atmosphere at the temperature of 140 °C, 280 °C, 480 °C and 580 °C, respectively. EC1 to EC3 are EC evolved in 98% helium/2% oxygen atmosphere at 580 °C, 740 °C and 840 °C, respectively. The red bar is the boundary of OC and EC, and the numbers show the mass percentage of OC in TC. OP is indicated as the grey area below the red bar, and is the difference between OC and OC1 + OC2 + OC3 + OC4. EC1-OP is indicated as the grey area above the red bar (EC = EC1-OP + EC2 + EC3).

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3.3. Annual emissions from residential coal combustion and agricultural biomass burning

By combining $PM_{2.5}$, OC and EC EFs in Table 2 with nation-wide fuel consumption, $PM_{2.5}$, OC and EC emissions for 2012 were 757 Gg, 237 Gg and 343 Gg from residential coal combustion and 1238 Gg, 524 Gg and 37 Gg from residential agricultural biomass burning (Table 4). There annual estimates are comparable to estimates for 2005 from Lei et al. (2011) for residential coal combustion, but they are ~3–16 times lower for the residential biofuel burning. OC and EC emissions (sum of residential coal combustion and agricultural biomass) in this study are 3.7 and 2.5 times lower than 2010 estimates from Lu et al. (2011), respectively. Differences among these estimates are due to both EFs and fuel consumption amounts.

Geographical distributions of residential burning $PM_{2.5}$ emissions based on provincial populations for 2012 are shown in Fig. 2, with provincial per capita emission estimates summarized in Tables S3 and S4. OC and EC emissions had similar geographical distributions (Figs. S3 and S4), with the highest coal emissions found in Shanxi, Inner Mongolia, Henan, Hebei and Guizhou provinces and the highest biomass emissions found in Heilongjiang, Hubei and Sichuan.

The spatial variability of the emissions from these two energy sources reflects the geographical patterns in both fuel consumption and production. Shanxi, Inner Mongolia and Guizhou are China's major coal-producing regions (NBS, 2014b). As a result of coal availability, attractive prices and old winters, coal became the main household fuel. Pollution levels are also high in these provinces (Li et al., 2015). Provinces with large rural populations, such as Henan Province, both coal and agricultural biomass will continue to be burned for domestic purposes, and high emissions from both types of fuels will be the result.

4. Conclusions

$PM_{2.5}$, OC and EC EFs from the residential combustion of bituminous coal, anthracite coal and agricultural biomass (wheat straw, maize straw and rice straw) in China were quantified in laboratory simulations, which will lead to a more quantitative understanding of the emission characteristics in residential sector source in China. These EFs were within the large range of values reported by other studies, with a significant relationship between the volatile matter content and EFs for coal combustion. The analyses of carbon fractions showed that (1) EC1-OP was the major fraction emitted from the burning of bituminous coal, (2) OC2 and OC3 were the main emissions from anthracite coal, and (3) OC2, OC3 and EC1-OP were the main products of agricultural biomass burning. According to the EFs measurements made in this study, it will be helpful to improve accurate and representative emission inventory in China based on their localization. Primary residential coal combustion $PM_{2.5}$, OC and EC emissions in China for 2012 were estimated at 757 Gg for $PM_{2.5}$, 237 Gg for OC and 343 Gg for EC while those for residential agricultural biomass burning were 1238 Gg, 524 Gg and 37 Gg of these species, respectively. Shanxi, Inner Mongolia, Henan, Hebei and Guizhou Provinces were the major sources of residential coal emissions while Heilongjiang, Hubei and Sichuan Provinces contributed the most in terms of agricultural biomass burning emissions.

Table 4
Comparison of Chinese $PM_{2.5}$, OC and EC emissions from residential coal combustion and agricultural biomass burning (units: Gg yr^{-1}).

Reference	Sources[a]	Emission			Year
		$PM_{2.5}$	OC	EC	
This work	RC/RB[b]	757/1238	237/524	343/37	2012
Lu et al. (2011)	RC + RB		2790	936	2010
Lei et al. (2011)	RC/RB	790/3600	320/2290	110/590	2005

[a] RC = residential coal combustion; RB = residential biofuel burning (including agriculture biomass, wood and other biofuel).
[b] Agricultural biomass only.

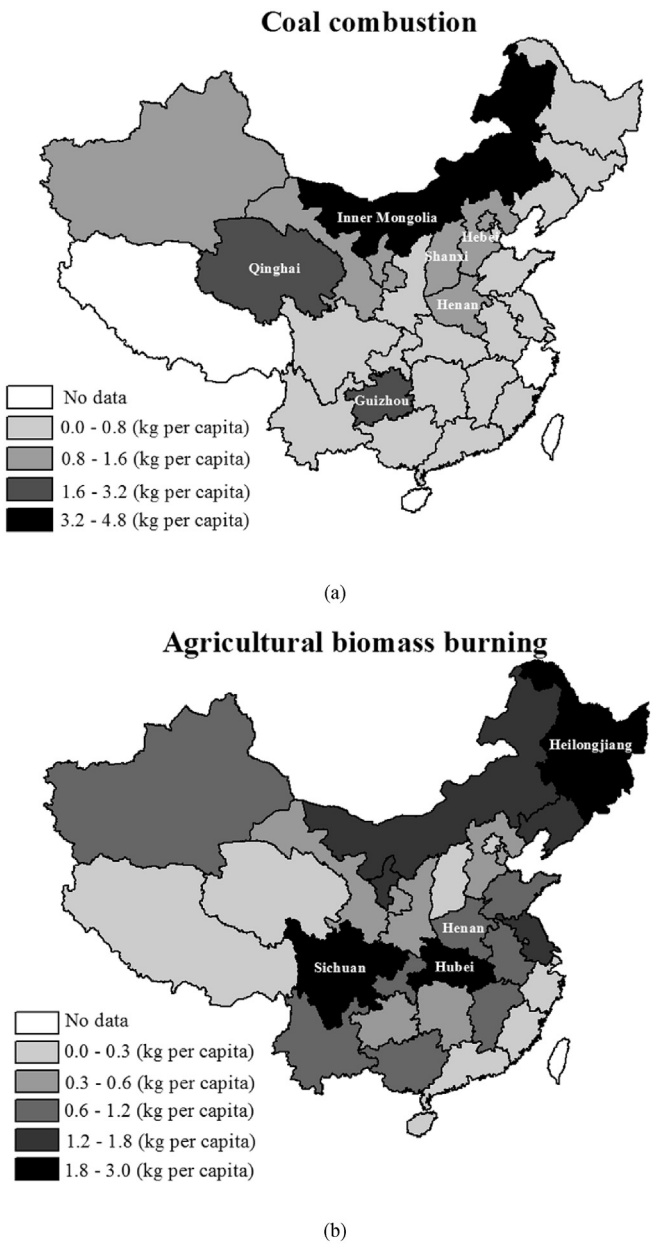

Fig. 2. Province-scale distribution in China of $PM_{2.5}$ per capita emissions from residential sources combustion in 2012: (a) coal combustion, and (b) agricultural biomass burning.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2016.12.006.

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