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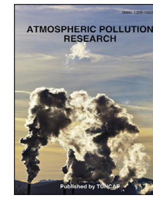

Characteristics of carbonaceous particles from residential coal combustion and agricultural biomass burning in China

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A B S T R A C T

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.

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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 $\mu g\ m^{-3}$, ~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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