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Emission factors of polycyclic and nitro-polycyclic aromatic hydrocarbons from residential combustion of coal and crop residue pellets[☆]

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) and nitro-polycyclic aromatic hydrocarbons (NPAHs) are toxic pollutants mainly produced during fossil fuel combustion. Domestic coal stoves, which emit large amounts of PAHs and NPAHs, are widely used in the Chinese countryside. In this study, emission factors (Efs) for 13 PAH species and 21 NPAH species for four raw coal (three bituminous and one anthracite), one honeycomb briquette, and one crop residue pellet (peanut hulls) samples burned in a typical Chinese rural cooking stove were determined experimentally. The PAH and NPAH Efs for the six fuels were 3.15–49 mg/kg and 0.32–100 µg/kg, respectively. Peanut hulls had very high Efs for both PAHs and NPAHs, and honeycomb briquettes had the lowest Efs. 2-Nitropyrene and 2-nitrofluoranthene, which are NPAHs typically found in secondary organic aerosol, were detected in the emissions from some fuels, suggesting that chemical reactions may have occurred in the dilution tunnel between the flue gas leaving the stove and entering the sampler. The 1-nitropyrene to pyrene diagnostic ratios for coal and peanut hulls were 0.0001 ± 0.0001 and 0.0005 , respectively. These were in the same order of magnitude as reference ratios for emissions during coal combustion. The 6-nitrobenzo[*a*]pyrene to benzo[*a*]pyrene ratios for the fuels were determined, and the ratios for coal and peanut hulls were 0.0010 ± 0.0001 and 0.0014 , respectively. The calculated potential toxic risks indicated that peanut hull emissions were very toxic, especially in terms of NPAHs, compared with emissions from the other fuels.

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1. Introduction

Attention is increasingly being paid to the presence of polycyclic aromatic hydrocarbons (PAHs) and nitro-polycyclic aromatic hydrocarbons (NPAHs), which are carcinogenic and/or mutagenic chemicals, in the environment (Ames et al., 1975; Epstein et al., 1979; Yang et al., 2010). PAHs and NPAHs in the atmosphere are mainly supplied through the incomplete combustion of fossil fuels and biomass (Shen et al., 2012; Yang et al., 2010). PAHs and NPAHs have been found in both the particulate and gaseous phases of combustion emissions and the ambient

atmosphere (Araki et al., 2009; Keyte et al., 2013; Masclat et al., 1986). Some NPAHs, such as 2-nitropyrene (2-NP) and 2-nitrofluoranthene (2-NFR), are produced in gas-phase reactions between the parent PAHs and hydroxyl (OH) and nitrate (NO₃) radicals (Arey et al., 1986). PAHs and NPAHs are semi-volatile, and PAH and NPAH partitioning between the particulate and gas phases has been widely studied. Most four- to six-ring NPAHs have been detected in the particulate phase (Araki et al., 2009; Bamford and Baker, 2003; Keyte et al., 2013).

Fossil fuel and biomass combustion emissions have been investigated intensively (De Gouw et al., 2004; Hayakawa et al., 1997; Li et al., 2009; Masclat et al., 1986; Schauer et al., 1999; Singh et al., 2013; Yang et al., 2010; Yokelson et al., 2009). China currently uses more coal than any other country, Chinese coal consumption in 2010 being 3.12×10^9 t (China Statistics Press, 2011). Crop residue combustion is also a major contributor to

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total energy consumption in China, next only to coal combustion, especially in rural areas in northern China (e.g., Huantai County, Shandong Province) (Zhou et al., 2009). Guo et al. (2012) estimated that 88.62 Mt of crop residues were combusted in China each year between 2007 and 2009, equivalent (using the calorific values of different types of biomass fuel) to 49.34 Mt of standard coal. During the same period, the amounts of peanut hull, cotton seed hull, sugarcane bagasse, and sugarbeet bagasse processing residues produced in China were 3.78×10^6 , 5.49×10^6 , 1.88×10^7 , and 4.40×10^5 t, respectively. Residential energy consumption (equivalent to 9.16×10^7 t of coal) is the second biggest contributor to total energy consumption in China (China Statistics Press, 2012). Combustion in domestic coal stoves for heating and cooking is widespread in suburban and rural areas in northern China, and crop residues are often used instead of coal as fuel (Chen et al., 2004a, 2004b, 2005; Liu et al., 2009; Zhou et al., 2009). A typical Chinese domestic stove is a cube or cylinder made mostly of cast iron. The stove body is usually 50–70 cm high, and the internal furnace typically has a volume of 5–7 L. Very serious PAH and NPAH pollution is caused by emissions from domestic stoves in which coal and biofuel are used (Chen et al., 2006; Shen et al., 2012; Zhi et al., 2008). Indoor combustion of firewood, indoor combustion of crop residues, and residential and commercial combustion of fossil fuel contributed 30.3%, 24.1%, and 7.6% of total PAH emissions, respectively, in China in 2007, according to emission estimates published by Shen et al. (2013b).

The relative contributions of PAHs and NPAHs from different sources of emissions are usually calculated using fuel consumption data and emission factors (Efs), which are usually affected by the fuel type, combustion apparatus, and sampling method. Some studies of PAH Efs for domestic coal and biomass combustion have previously been published (Chen et al., 2004a, 2004b, 2005; Shen et al., 2010; Singh et al., 2013; Tao et al., 2006; Xu et al., 2006; Yang et al., 2010), but biomass combustion in domestic stoves in China has been studied only a few times (Shen G. et al., 2011, 2012, 2013). Furthermore, few studies have been focused on NPAH Efs for coal and biomass combustion, and, in these studies, few NPAH species were investigated (Shen et al., 2012, 2013a). 1,3-Dinitropyrene (DNP), 1,6-DNP, and 1,8-DNP are several to 2000 times more mutagenic than benzo[*a*]pyrene (Rosenkranz and Mermelstein, 1983), and they are considered to be “possible human carcinogens” (Group 2B) by the World Health Organization (2017). We have previously described the characteristics of NPAHs, including the three DNPs mentioned above, in domestic coal stove emissions (Tang et al., 2005; Yang et al., 2010). However, we only tested a small number of fuels at that time. Furthermore, a dilution tunnel was not used in the burning and sampling system in those studies, so the NPAH Efs we calculated could not be converted to fuel-weight-based values and only particulate-weight-based Efs could be assessed.

In the study described here, a combustion experiment was performed using a typical domestic stove and experimental conditions matching actual residential burning practices. Four domestic coal samples and one crop residue pellet sample were tested because these are typical fuels used in residential stoves in China. Part of one of the coal samples was converted into a honeycomb-coal-briquette type sample and tested for comparative purposes. We determined 13 PAHs and 23 NPAHs in the emission samples collected when the six fuel samples were combusted. The analyses were performed by high performance liquid chromatography (HPLC). The aim was to investigate the characteristics of the PAH and NPAH emissions (the Efs, gas–particulate distributions, and potential toxic risks) when the fuels were combusted in a domestic stove.

2. Experimental

2.1. Fuels and stove

Four raw coal (three bituminous and one anthracite) samples and one crop residue pellet sample were selected for use in the emission tests. A portion of one of the coal samples was converted into honeycomb briquettes and also used in the tests. The bituminous coal samples were collected from Yinchuan (YC, Ningxia Province), Dongsheng (DS, Inner Mongolia), and Datong (DT, Shanxi Province), and the anthracite was collected from Zhijin (ZJ, Guizhou Province). The coals were cut into 5–8 cm blocks for use in the combustion tests. DT coals were converted into honeycomb briquettes (later called DT-HB) by grinding the coal and mixing the powder with clay using an automated machine. Each briquette was 6 cm high and 0.5 cm in diameter, with 12 columns of holes. The biomass fuel that was tested was peanut hull pellets, purchased from a local market. The pellets were block-formed and approximately 3.2 cm × 3.2 cm × (5–8) cm. A typical domestic coal-stove was purchased from a local market for use in the combustion tests. The stove had a grate for the fire to burn on, and the ash was collected on a removable iron drawer beneath the grate. Each fuel sample was analyzed by the China Coal Research Institute using Chinese national standard method GB/T212-2008 (General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China, 2008), and the properties of the samples are shown in Table 1.

2.2. Sampling system

As shown in Fig. 1, coal combustion flue gas entered the dilution tunnel through a small stack over the stove. The dilution tunnel had an orthogonal pipe and a horizontal cylindrical tunnel, both made of polished stainless steel. The orthogonal pipe was connected to the stove and allowed the flue gas to be introduced to the dilution tunnel. The flue gas was diluted with filtered air a first time in the orthogonal pipe. The horizontal cylindrical tunnel was connected to the opposite end of the orthogonal pipe. A second dilution step occurred in the horizontal tunnel. There were some outlets at the end of the tunnel to allow the main pump to be connected and samples to be collected. The main pump flow rate was 6000 L/min. The residence time of the exhaust gas in the dilution tunnel was 5.5 s. After the dilution steps, the flue gas temperature had decreased to about 30 °C. Two thermometers were fitted to the system, one to measure the temperature inside the furnace and the other to measure the temperature of the diluted flue gas.

Particulate-bound PAHs and NPAHs in the flue gases produced in each test were sampled by collecting a particulate sample on a 2500QAT-UP quartz fiber filter (90 mm diameter; Pallflex Products, Putnam, CT, USA) fitted in a Dickel-80 mid-volume particulate sampler (Beijing Geological Instrument-Dickel Cooperation Limited, Beijing, China) using a flow rate of 78 mL/min. The gaseous PAHs were collected on XAD-4 resin packed in a column with polyurethane foam at each end. The column was connected directly under the filter described above. The XAD-4 resin (diameter 250–800 μm, pore size 48 Å, volume/weight ratio 0.96 cm³/g, surface area 725 m²/g) was purchased from Rohm and Haas (Philadelphia, PA, USA).

2.3. Sample collection

Entire residential combustion practice cycles were simulated in the experiments. At the beginning of a combustion test, about 0.5 kg charcoal was ignited in the stove. The test fuel was added to

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