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Polycyclic aromatic hydrocarbon emission from straw burning and the influence of combustion parameters

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ABSTRACT

A simulated burning experiment was conducted in a tubular furnace system to examine the emission of polycyclic aromatic hydrocarbons (PAHs) from the burning of rice and bean straw, and the influence of combustion parameters was investigated. Total emission amounts of 16 PAHs (∑PAHs) from the burning of rice and bean straw ranged from 9.29 to 23.6 μg g^{-1} and from 3.13 to 49.9 μg g^{-1}, respectively, which increased with the increase of temperatures from 200 to 700 °C. The contribution of combustion to individual PAH yields was about 80.6–100%, which was generally increased with the increase of burning temperature. Moisture content in straw had a negative effect on PAH formation, especially on PAHs with low molecular weight. ∑PAHs emission amounts decreased by 78.2% for bean straw with a moisture content of 30% in comparison with that for dried straw. In addition, PAH emission amounts increased with the increase of O_2 content in supplied air and then decreased, which showed a maximum emission at O_2 content of 40%. The source fingerprint of PAHs in emission from straw burning was established, which showed that naphthalene accounted for 35.0 ± 7.4% of ∑PAHs. Based on the experimental data, emission amounts of ∑PAHs from the burning of rice and bean straw were estimated to be 320–357 and 32.5–76.0 tons to ambient air per year in China, respectively.

1. Introduction

Biomass burning has induced global concerns in the past decades for its effects on visibility, human health and global climate by emitting particles and vapor pollutants (Fang et al., 1999; Koe et al., 2001; Radzi bin Abas et al., 2004; Yang et al., 2006). Open burning of biomass is a common method for agricultural residue disposal and represents a considerable source of atmospheric pollutants (Jenkins et al., 1996; Korenaga et al., 2001). Principal pollutants emitted are CO, hydrocarbons and particles, with smaller amounts of NO_x and SO_2. In addition, there are emissions of volatile organic compounds and polycyclic aromatic hydrocarbons (PAHs), many of which have carcinogenic properties to humans (Amagai et al., 1999; Liu et al., 2001; Ohura et al., 2004a).

PAHs are widespread environmental pollutants, typically containing two to eight aromatic rings, and formed during incomplete combustion of organic materials (Terzi and Samara, 2005; Lu and Zhu, 2007). The current understanding of the effects of combustion parameters on PAH emission during biomass combustion is at an early stage. Very few studies have been carried out to identify the effects of combustion parameters on PAH emission other than temperature (Fängmark et al., 1993; Gulyurtlu et al., 2003). Even in the case of temperature, the reported results were conflicting. Therefore, a detailed investigation is needed to identify the influence of combustion parameters on PAH emission from biomass burning.

In China, rice is the predominant crop in the south, and bean is widely planted in the north. Thus, rice and bean straws are two important species of biomass in China. The availability of these straws amounted to 132 million tons in China, of which 29.3 million tons were burnt in 2003 (Cao et al., 2006). In this work, rice and bean straws were selected to examine the PAH emission from their burning in a tubular furnace system. The objectives of this work were: (1) to assess PAH emission levels from the burning of rice and bean straw; (2) to investigate the influence of combustion parameters, including temperature, moisture content and O_2 content in supplied air, on PAH emission from straw burning; (3) to establish the fingerprint of PAH emission from straw burning and provide the data to evaluate PAH emission amounts from straw burning in China.

2. Experimental methods

2.1. Burning system

A schematic view of the tubular furnace system is shown in Fig. 1. The system consisted of three parts: gas supply, tubular

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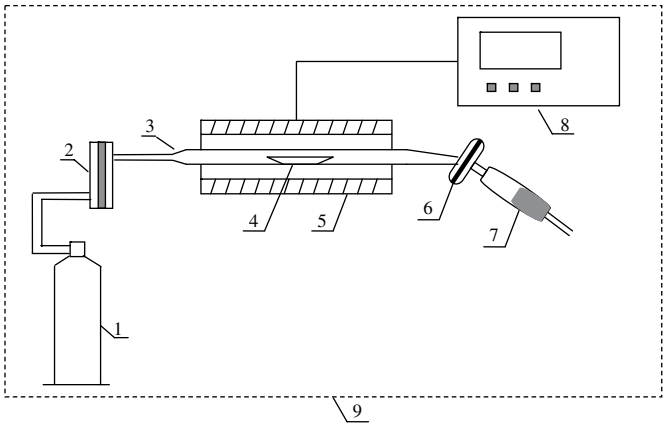

1. Gas tank; 2. Flow meter; 3. Quartz tube; 4. Ceramic boat; 5. Heater;
6. Glass fiber filter; 7. XAD-2 resin; 8. Temperature control panel;
9. Ventilation cabinet

Fig. 1. Schematic view of the tubular furnace system.

furnace and PAH sampling. At the beginning of the experiment, the tubular furnace was set to a certain temperature, and the gas flowed into the quartz tube at a rate of 1.0 L min^{-1}. Then, the ceramic boat containing straws was quickly put into the quartz tube, and the PAH sampling tube was simultaneously connected to the quartz tube. PAHs in the smoke from straw burning were collected by XAD-2 resin and glass fiber filters.

2.2. Preparation of straw samples and combustion parameters

The pulverized rice and bean straws were oven dried at 105 °C for 24 h and then placed in a desiccator to reach room temperature before loading with moisture. Straws (0.6 g) were placed in the ceramic boat and burned for 20 min in each test. The tests were repeated three times, and the results shown in this paper are average values.

The moisture content in dried rice and bean straws was considered to be zero, and they were burned at 200, 300, 400, 500, 600 and 700 °C with standard air ($V_{N2}/V_{O2} = 79/21$) supplied, respectively.

Bean straws with different moisture contents were prepared by sealing 20 g dried straws with different amounts of water in vinyl bags, and shaking the bags until the water was absorbed completely. The moisture content of the straws was 0, 10, 20 and 30%, respectively. The samples were burned at 700 °C with standard air supplied.

Dried bean straw was also burned at 700 °C with different gas mixtures (N_2 and O_2, Jingong, China) supplied. O_2 content accounted for 0, 21, 40, 60, 80 and 100% of mixed gas, respectively.

2.3. Analysis of PAH samples

Vapor phase PAHs were adsorbed by XAD-2 resin (2.5 g, Supelco, USA), which was cleaned with dichloromethane and methanol until peak of PAHs could not be found in HPLC. Particulate PAHs were collected with 25 mm glass fiber filters (GF, Whatman, England), which were thermally treated at 500 °C for 6 h to remove organic contaminants. After sampling, the glass fiber filters were cut into pieces, and placed in a 25 mL glass stoppered tube with 10 mL dichloromethane. The XAD-2 resin was poured into a 25 mL glass stoppered tube with 20 mL of a mixture of dichloromethane and acetonitrile (V/V = 3/2). Then the samples were sonicated for 30 min. During the sonication, the water in the ultrasonic bath was replaced frequently to prevent overheating. Then 2 mL extracts of XAD-2 resin and glass fiber filters were transferred through a 2.0 silica gel column with 11 mL of a solution of hexane and dichloromethane mixture (V/V = 1/1), respectively. The solutions with 50 μL of dimethyl sulfoxide (DMSO) were evaporated under a gentle flow of N_2 gas at room temperature and then 950 μL of acetonitrile were added.

About 0.2 g of dried straw and burning residue were placed in a 25 mL glass stoppered tube with 10 mL dichloromethane and sonicated for 30 min, respectively. The subsequent steps were the same as above.

The samples were analyzed for the following 16 PAHs: naphthalene (NA), acenaphthylene (ACY), acenaphthene (AC), fluorene (FLUOR), phenanthrene (PHEN), anthracene (AN), fluoranthene (FLUR), pyrene (PY), benzo[a]anthracene (BaA), chrysene (CHRY), benzo[b]fluoranthrene (BbF), benzo[k]fluoranthrene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DA), benzo[ghi]perylene (BP), indeno[1,2,3-cd]pyrene (IN). The PAHs were determined by HPLC (Agilent 1200, USA) containing a LiChrospher PAH column (250 × 4.6 mm, Agilent, USA), a fluorescence detector and an ultraviolet detector (to determine ACY only). Sixteen PAHs were separated in HPLC with linear gradient mobile phases of water and acetonitrile (V/V was maintained at 60/40 from 0 to 3 min, and charged to 0/100 from 3 to 30 min, then kept at 0/100 from 30 to 42 min). The flow rate of mobile phases was 1.0 mL min^{-1}, and the column was maintained at 30 °C.

2.4. Quality control

PAH recovery studies ($n = 5$) were undertaken to demonstrate the availability of the analytical method. We produced cleaned XAD-2, glass fiber filters and nothing doped with PAH standard samples (Supelco, USA) to correspond to concentrations in extracts of, for example, 1, 2, 3, 4, 5 μg mL^{-1} in the vapor phase, 0.5, 1, 1.5, 2, 2.5 μg mL^{-1} in the particulate phase, and 0.02, 0.04, 0.06, 0.08, 0.1 μg mL^{-1} in the blank sample for PHEN, respectively. The recoveries of 16 PAHs ranged from 85.2 ± 2.0% to 98.7 ± 3.8% except for NA (its recoveries were 77.0 ± 2.7%, 78.8 ± 5.4%, 78.8 ± 5.4% for vapor phase, particulate phase, straw and burning residue, respectively, Table 1).

3. Results and discussion

3.1. PAH emission from dried straw burning

The furnace temperature varied from 200 to 700 °C with intervals of 100 °C, because the actual temperature would not exceed this scope during open straw burning (Lu et al., 2005). The results of

Table 1
The recoveries of 16 PAHs (%).

	Vapor	Particulate	Straw and burning residue
NA	77.0 ± 2.7	78.8 ± 5.4	75.9 ± 3.5
ACY	86.4 ± 2.6	87.4 ± 5.0	85.9 ± 3.6
AC	85.7 ± 4.2	87.2 ± 3.7	87.9 ± 4.3
FLOUR	88.0 ± 6.2	85.3 ± 4.9	86.1 ± 4.6
PHEN	98.4 ± 4.2	96.5 ± 5.0	97.5 ± 2.7
AN	89.9 ± 3.0	91.2 ± 3.4	85.6 ± 2.9
FLUR	94.5 ± 4.0	94.2 ± 4.7	96.1 ± 3.1
PY	87.9 ± 1.6	91.1 ± 4.9	98.4 ± 2.6
BaA	91.5 ± 3.2	92.6 ± 4.6	94.1 ± 2.8
CHRY	92.6 ± 3.3	88.7 ± 4.5	95.4 ± 3.8
BbF	93.8 ± 3.5	90.4 ± 5.5	94.8 ± 2.4
BkF	94.8 ± 2.5	94.6 ± 2.7	94.7 ± 2.5
BaP	95.7 ± 3.4	96.2 ± 2.6	87.1 ± 2.4
DA	93.5 ± 4.6	93.1 ± 4.4	91.2 ± 4.2
BP	98.7 ± 3.8	94.9 ± 3.2	86.7 ± 3.4
IN	96.2 ± 4.3	90.9 ± 2.2	85.2 ± 2.0