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Emission and size distribution of particle-bound polycyclic aromatic hydrocarbons from residential wood combustion in rural China

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ARTICLE INFO

Article history:

Received 27 July 2012

Received in revised form

8 January 2013

Accepted 27 January 2013

Available online 7 March 2013

Keywords:

Polycyclic aromatic hydrocarbon

Residential wood combustion

Emission factor

Size distribution

Cooking stove

ABSTRACT

Emissions and size distributions of 28 particle-bound polycyclic aromatic hydrocarbons (PAHs) from residential combustion of 19 fuels in a domestic cooking stove in rural China were studied. Measured emission factors of total particle-bound PAHs were 1.79 ± 1.55 , 12.1 ± 9.1 , and 5.36 ± 4.46 mg kg⁻¹ for fuel wood, brushwood, and bamboo, respectively. Approximately 86.7, 65.0, and 79.7% of the PAHs were associated with fine particulate matter with size less than 2.1 μm for these three types of fuels, respectively. Statistically significant differences in emission factors and size distributions of particle-bound PAHs between fuel wood and brushwood were observed, with the former had lower emission factors but more PAHs in finer PM. Mass fraction of the fine particles associated PAHs was found to be positively correlated with fuel density and moisture, and negatively correlated with combustion efficiency. Low and high molecular weight PAHs preferably segregated into the coarse and fine PM, respectively. The high accumulation tendency of the PAHs from residential wood combustion in fine particles implies strong adverse health impact.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), a group of pollutants mainly produced from incomplete combustion, are well known for their mutagenic and carcinogenic potential and are among the toxic organics of growing concern in many countries. Personal exposure to PAHs was suggested to associate with the increased risk of lung cancer [1,2]. It was estimated that exposure to ambient air PAHs contributed to approximately 1.6% of lung cancer cases in China [2].

Total global PAH emission was estimated to be 520 Gg in 2004, among which 80% was from developing countries and 57% was from biomass burning [3]. Emission of such a large amount of PAHs into the environment results in severe contamination at both regional and global scales. Ubiquitous PAH pollution in various environmental media and foodstuffs has been reported in the literature [4–8]. In China, the pollution was much more serious due to higher emissions from incomplete combustions under lower burning efficiencies [9–11].

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<http://dx.doi.org/10.1016/j.biombioe.2013.01.031>

PAHs in ambient air or emission smoke are in either gaseous or particulate phase [4,12], and usually absorption and/or adsorption to particulate matter (PM) were the main governance mechanism(s) of particle-bound PAHs [13–15]. The health effects of PM are strongly size dependent [13,16]. Different effects of coarse, fine and ultra-fine PM on respiratory and cardiovascular diseases have been documented in both laboratory toxicology experiments and epidemiological studies [17–21]. Though the mechanism of PM induced toxicity was not fully understood, chemical composition, including metals and toxic organics, like PAHs, is recognized as one of the key factors. It is also expected that PAHs in PM of different size cause distinct toxic effects, and PAHs in finer PM may produce higher toxic effects since fine particles can penetrate deep into the lung region [19].

Residential wood combustion (RWC) is one of the major sources of PAHs, especially in developing countries due to large consumption and low combustion efficiency [22–24]. Primary PM from RWC is fine and usually has a large fraction of carbonaceous soot with high levels of organics including PAHs [25–27]. In China, RWC contributed 20–35% of the total PAH emissions [28,29]. Emissions of various pollutants from residential wood combustion have been extensively investigated in the previous studies, which developed effective research methods for the emission characterization of RWC, and reported a large number of valuable data on the emission factors of many incomplete pollutants from different fuel/stove combinations under varied burning conditions [30–33]. However, these studies were mainly conducted in U.S. or European countries where fuel types and stove designs are rather different from those used in rural China. In addition, most of the current studies were done in laboratory. It is believed that emissions from the field differ from those in laboratory chamber studies [34]. Moreover, emissions of PAHs in PM with different diameters were rarely characterized [13,15], which prevents us from full understanding the health effects of these RWC emitted PM and PAHs.

In this study, size resolved particle-bound PAHs from RWC in a typical cooking stove were measured in a rural kitchen. The impacts of fuel property and combustion efficiency on the size distributions of particulate phase PAHs were investigated, and composition profiles of PAHs in PM of different diameters are discussed.

2. Material and methods

2.1. Fuel combustion experiments

Nineteen biomass fuels, including 15 fuel wood (Chinese white poplar (*Populus tomentosa* Carr.), water Chinese fir (*Metasequoia glyptostroboides*), Chinese pine (*Pinus tabulaeformis* Carr.), cypress (*Cupressus funebris* Endl.), elm (*Ulmus pumila* L.), fir (*Cunninghamia lanceolata*), larch (*Larix gmelini* (Rupr.) Rupr.), maple (*Acer mono* Maxim.), paulownia tomentosa (*P. tomentosa* (Thunb.) Steud.), willow (*Salix babylonica*), locust (*Robinia pseudoacacia* L.), ribbed birch (*Betula dahurica* Pall.), paulownia elongata (*P. elongata* S. Y. Hu), black poplar (*Populus nigra* L.), and aspen (*Populus adenopoda* Maxim.), 3 brushwood (*Lespedeza bicolor* Turcz), holly (*Buxus megistophylla* Lévl), and

buxus sinica shrub (*Buxus sinica* (Rehd. et Wils.) Cheng)), and 1 bamboo (*Phyllostachys heterocycla*(Carr.)) were burned in a typical brick stove. The wok stove was 0.80 m in length, 0.70 m in width and 0.65 m in height, with one iron pot in the middle. The chamber volume was approximately 0.20 m³. The grate to the pot bottom distance and grate to the ground distance were 0.30 and 0.15 m, respectively. A picture of the wok stove is shown in Figure A.1. Pre-weighed wood pieces (10–15 cm² × 10–20 cm in length) were ignited and inserted into the stove in 15–20 batches. Each wood type was burned separately. Triplicate burning experiments were conducted for each fuel. The sampling covered the whole burning period lasting for 40–60 min. The exhaust from the stove chamber passed through a heating bed (known as “Kang” in Chinese) and then entered into a mixing chamber (4.5 m³), where a fan was installed and on all the time to mix the exit smoke and minimize the influence of temperature on sampling [35]. The samples were collected directly from the chamber and no dilution was conducted to avoid the potential impacts of dilution rates and ratios on mass load and size distribution [34,36]. Smoke temperature and relative humidity measured in the mixing chamber were in the range of 293–318 K and 40–65%, respectively (TM184, Tenmars). It is believed that significant condensation of water vapor could not occur at this condition [34]. Experimental set-up and combustion procedure were same as those reported previously on EFs of carbonaceous PM from residential biomass burning [35,37]. The measured fuel properties, including density, moisture, elemental contents and proximate analysis results, for the 19 fuels investigated are provided in Table A.1.

2.2. Sample collection and extraction

Sampling collection and extraction methods followed the procedure in a previous study on PAH emission from indoor crop residue burning [38] with a small modification. Particle bound PAHs were collected on glass fiber filters and 4 size segregated samples with PM diameters of <0.4, 0.4–1.1, 1.1–2.1, and 2.1–10 µm were collected using a cascade impactor at a flow rate of 28.3 dm³ min^{−1} (FA-3, Kangjie, China).

Particle-bound PAHs were extracted using a microwave accelerated system (CEM Mars Xpress, USA) Microwave power was set at 1200 W (100%). The temperature program was to 383 K in 10 min and then held for another 10 min. After extraction, the extracts were concentrated to 1 cm³ and transferred to a silica/alumina gel column for cleanup. The column was packed with 12 cm alumina, 12 cm silica gel, and 1 cm anhydrous sodium sulfate from bottom up, and pre-eluted with 20 cm³ hexane. Target PAHs were eluted with 70 cm³ hexane/dichloromethane (1:1) mixture. The eluate was finally concentrated to 1 cm³ and spiked with deuterated internal standards (J&W Chemical, USA).

2.3. PAH analysis and quality control

PAH analysis was performed using a gas chromatograph (GC, Agilent 6890) connected to a mass spectrometer (MS, Agilent 5973) in electron ionization mode. An HP-5MS capillary column (30 m × 0.25 mm × 0.25 µm) was used. The oven

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