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Short Communication: Emission of Oxygenated Polycyclic Aromatic Hydrocarbons from Biomass Pellet Burning in a Modern Burner for Cooking in China

Guofeng Shen, Siye Wei, Yanyan Zhang, Rong Wang, Bin Wang, Wei Li, Huizhong Shen, Ye Huang, Yuanchen Chen, Han Chen, Wen Wei, and Shu Tao*

Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

Abstract

Biomass pellets are undergoing fast deployment widely in the world, including China. To this stage, there were limited studies on the emissions of various organic pollutants from the burning of those pellets. In addition to parent polycyclic aromatic hydrocarbons, oxygenated PAHs (oPAHs) have been received increased concerns. In this study, emission factors of oPAHs (EF_{oPAHs}) were measured for two types of pellets made from corn straw and pine wood, respectively. Two combustion modes with (mode II) and without (mode I) secondary side air supply in a modern pellet burner were investigated. For the purpose of comparison, EF_{oPAHs} for raw fuels combusted in a traditional cooking stove were also measured. EF_{oPAHs} were 348 ± 305 and 396 ± 387 $\mu\text{g}/\text{kg}$ in the combustion mode II for pine wood and corn straw pellets, respectively. In mode I, measured EF_{oPAHs} were 77.7 ± 49.4 and 189 ± 118 $\mu\text{g}/\text{kg}$, respectively. EFs in mode II were higher (2–5 times) than those in mode I mainly due to the decreased combustion temperature under more excess air. Compared to EF_{oPAHs} for raw corn straw and pine wood burned in a traditional cooking stove, total EF_{oPAHs} for the pellets in mode I were significantly lower ($p < 0.05$), likely due to increased combustion efficiencies and change in fuel properties. However, the difference between raw biomass fuels and the pellets burned in mode II was not statistically significant. Taking both the increased thermal efficiencies and decreased EFs into consideration, substantial reduction in oPAH emission can be expected if the biomass pellets can be extensively used by rural residents.

Keywords

Oxygenated PAHs; Biomass Pellets; Emission Factor; Emission Reduction

*Corresponding author phone and fax: 0086-10-62751938, taos@pku.edu.cn.

Appendix. Supplementary material

Supplementary material associated with this article including the experimental outline, pictures of the pellets and stoves, laboratory analysis procedure of oPAHs, calculation of emission factor based on carbon mass balance method, composition profiles and emission reduction calculation, can be found at the website free of charge.

1. Introduction

Oxygenated polycyclic aromatic hydrocarbons (oPAHs) which have one or more oxygen(s) attached to the aromatic rings of their parent polycyclic aromatic hydrocarbons (pPAHs) are of growing concern due to their abundance and persistence in environment, and more direct toxic potential compared to the pPAHs (Bolton et al., 2000; Lundstedt et al., 2007; Walgraeve et al., 2010). Polar oPAHs usually have relatively higher molecular weights, lower vapor pressures, and subsequently higher tendencies to be associated with fine particulate matters than the corresponding pPAHs (Walgraeve et al., 2010). It is believed that oPAHs are related to the generation of reactive oxygen species which can enhance the oxidative stress in cells, formation of protein and DNA adducts, depletion of glutathione, and endocrine effects (Bolton et al., 2000; Humagai, 2009; Walgraeve et al., 2010).

The measured ambient concentrations of oPAHs (several pg/m^3 to ng/m^3) were generally comparable to those of pPAHs (Walgraeve et al., 2010). Among various oPAH found in environment, several ketones, like 9-fluorenone (9FO), benzanthrone (BZO), and quinones like anthrathrene-9,10-quinone (ATQ) and benz[a]anthracene -7,12-dione (BaAQ) are often the most abundant (Albinet et al., 2008; Aldreou et al., 2009; Allen et al., 1997; Fitzpatrick et al., 2007; Lundstedt et al., 2007).

oPAHs can be produced either from primary emissions or secondary formation through the reaction of parent aromatics with radicals. The predominance of the primary and/or secondary sources of oPAHs depends on compound, site, and environmental conditions (Eiguren-Fernandez et al., 2008; Kojima et al., 2010; Walgraeve et al., 2010). For the primary combustion sources, oPAHs were identified from indoor combustions of crop residues (Shen et al., 2011), wood (Gullett et al., 2003), coals (Shen et al., 2011) as well as open biomass burning (Hays et al., 2005) and diesel exhaust (Choudhury, 1982).

Biomass pellets, as a cleaner alternative of the raw biomass, have been deployed in the last several decades in many countries, like Sweden, United States, and also China (Boman et al., 2011; Johansson, et al., 2004; Olsson et al., 2003; Perzon, 2010). In Sweden, consumption of biomass pellets in residential sector increases at an annual rate of 30% (Olsson et al., 2003). According to the Medium and Long-Term Development Plan for Renewable Energy in China, use of biomass pellets is to be strongly supported, targeting at 50 million tons in 2020 (Chen et al., 2009). While the thermal efficiencies of pellet burners were well documented, there is limited information on the emission characterization of the pellet burning (Boman et al., 2011).

To the best of our knowledge, there is no measurement on the emission of oPAHs from biomass pellet burning. The objectives of this study were 1) to measure EFs of oPAHs (EF_{oPAHs}) from two types of pellets in a modern pellet burner; 2) to study the difference in EF_{oPAHs} from two combustion modes with and without secondary air supply; and 3) to address potential impact of pellet deployment on oPAH emissions from residential sector by comparing EF_{oPAHs} measured for pellets in a modern burner with those for raw biomass fuels burned in a brick cooking stove.

2. Material and methods

2.1 fuel, stove and combustion experiment

Two commercial biomass pellets made from corn straw (*Zea mays*) and pine wood (*Pinus tabulaeformis* Carr.), respectively, were combusted in a modern burner purchased from a local market in rural Beijing. The burner was a manually top fed burner with power of about 2.64 kW. The picture of the modern pellet burner, and the measured fuel properties including density, moisture, element contents and the result of proximate analysis are provided in Table S1. Biomass pellets had higher bulk densities, ash contents, but lower moisture and volatile matter contents than the corresponding raw fuels. Relatively high ash content of the pellets might be related to the contamination during handling and storage, or additives used (Wiinikka, 2005; Yao et al., 2010). A comparison study on the fuel properties of biomass pellets from China and Sweden showed that ash contents of the former were 7.71–21.7% and 1.01–9.25% for crop straw pellets and wood pellets, respectively, which were generally higher than those of 4.70–7.90% and 0.30–3.40% for Swedish pellets (Yao et al., 2010).

The combustion experiments were done in a rural kitchen (S2) where previous studies on emissions from residential solid fuel combustions were conducted (Shen et al., 2010, 2012). Pre-weighed pellets were added into the burner and fired. After ignition, the burner was set under a stainless hood and the existed smoke from the combustion entered into a mixing chamber (4.5 m³ with a small fan built-in) through a stainless pipe. There was no further dilution done to avoid the potential influence of dilution rate and ratio on PM mass load. All sampling and measurements were done in the mixing chamber. The secondary formation of oPAHs in the mixing chamber can not be totally ruled out. However, without flow rate measured in this study, residence time in the mixing chamber could not be calculated. The pellets were burned in two different modes without (mode I) and with (mode II) secondary side air supply (Figure S1), respectively.

For a comparison, emissions for raw corn straw and pine wood were also measured. A brick wok stove widely used in Northern China was used (Shen et al., 2010). Pre-weighed raw biomass fuels (the same quantities of those of the pellets) were inserted into the combustion stove in 8–10 batches, as residents do in daily lives. The exited smoke passed through a heating bed (known as “Kang” in Chinese) and entered into the mixing chamber. The burning lasted for 10–15 min. Sampling covered the whole burning cycle. The combustion experiments were repeated three times. Combustion temperature in the stove chamber was measured (TM 902C digital thermometer) and recorded every 3 minute during the processes. The temperature sensor was put in the center of the stove chamber. The detailed information of the stove and combustion procedure can be found in a previous published paper (Shen et al., 2010).

2.2 sampling, extraction and analysis

Sampling, extraction, and laboratory analysis procedure were the same as those in the previous study on the oPAH emissions from indoor solid fuel combustion (Shen et al., 2011). Particulate and gaseous phase samples were collected on the quartz fiber filters

(QFFs, 22 mm in diameter) and polyurethane form plugs (PUFs, Supelco, 22 mm diameter $\times$ 7.6 cm, 0.024 g/cm³), respectively. The PUFs were extracted using Soxhlet with 150 mL dichloromethane. QFFs were extracted using a microwave accelerated system (Mars Xpress, USA) with 25 hexane/acetone mixture. Target oPAHs were analyzed using a gas chromatograph (GC, Agilent 6890) coupled with a mass spectrometer (MS, Agilent 5975) equipped with a HP-5MS capillary column (30m $\times$ 0.25mm $\times$ 0.25 μ m) in chemical ionization mode. Detailed information about laboratory analysis and quality control is provided in the Supporting Information (S3).

EFs of oPAHs were calculated based on the carbon mass balance method (see Supporting Information-S4) assuming that the total carbon emitted from the fuel combustion presented in the forms of gaseous CO, CO₂, total hydrocarbon, and total carbon in particulate matter (Zhang et al., 2000). Statistica was applied for data analysis and a significance level of 0.05 was adopted.

3. Results and Discussion

3.1 EFs of oPAHs for the Biomass Pellets

The measured EFs of the 4 oPAHs for two types of pellets burned in two modes were listed in Table 1 as means and standard deviations. EFs of CO (EF_{CO}), a main product of incomplete combustion, were also provided. For pine wood pellets, total gaseous and particle-bound oPAHs were 43.9 $\pm$ 42.6 and 33.8 $\pm$ 16.9 μ g/kg in combustion mode I, and 92.6 $\pm$ 88.5 and 221 $\pm$ 225 μ g/kg in mode II, respectively. The EFs of total oPAHs in mode II (348 $\pm$ 305 μ g/kg) were significantly ($p < 0.05$) higher (a factor of 4.5) than those in mode I (77.7 $\pm$ 49.4 μ g/kg), likely due to the increased excess air and cooled temperature in combustion mode II when there were more air amounts supplied from the burner side (Boman et al., 2011; Johansson et al., 2004). The measured temperatures in the burner during the pine pellet burning were 550–750 and 650–900°C in mode II and mode I, respectively. EF_{CO} were 2.69 $\pm$ 0.43 and 6.07 $\pm$ 2.04 g/kg in mode I and II, respectively. EF_{CO} in mode II was also significantly higher ($p < 0.05$) than that in mode I, consistent with the difference in EF_{oPAHs}. In both EF_{CO} and EF_{oPAHs}, relative large variations existed due to the differences in fuel property and combustion conditions. The coefficients of variation (COVs) in EF_{CO} in mode I and II were 16 and 34%, and COVs in EF_{oPAHs} were 64 and 88% in two modes, respectively.

For the corn straw pellet, EFs of total gaseous and particle-bound oPAHs were 75.0 $\pm$ 86.1 and 114 $\pm$ 88.5 μ g/kg in mode I, and 87.6 $\pm$ 73.2 and 309 $\pm$ 320 μ g/kg in mode II, respectively. Total EF_{oPAHs} were 189 $\pm$ 118 and 396 $\pm$ 387 μ g/kg in combustion mode I and II, respectively. It appears that the relative large variation in EF measurements resulted in the insignificant difference ($p > 0.05$) between two modes, although the measured temperatures were also obviously lower in mode II (400–700°C) than those in mode I (500–800°C). EF_{CO} were 15.2 $\pm$ 8.3 and 27.1 $\pm$ 20.6 g/kg in mode I and II, respectively. Similar to EF_{oPAHs}, EF_{CO} in mode II was not significantly different from that in mode I ($p > 0.05$), though the later was obviously higher than the former. COVs in EF_{CO} for corn straw pellets were 55 and 76%, and were 62 and 98% in EF_{oPAHs} in combustion mode I and II, respectively.

There was no significant difference ($p > 0.05$) in EFs of oPAH individuals between the two types of pellets in the same combustion mode, except EFs of BZO (8.68 ± 8.03 and 68.0 ± 49.3 $\mu\text{g/kg}$) for pine wood and corn straw pellets in mode I. It is believed that EFs can be influenced by various factors including fuel properties (e.g. fuel moisture, volatile matter content, heating values) and combustion conditions (e.g. modified combustion efficiency (MCE, defined as $\text{CO}_2/(\text{CO}_2 + \text{CO})$) and burning rate) (Dhammapala et al., 2006; Johansson et al., 2004; McDonald et al., 2000; Shen et al., 2010).

For the two biomass pellets studied, pine wood pellets had higher volatile matter contents which can result in higher EFs. However, lower EFs from pine pellet burning could be expected since calculated MCEs for the burning of pine pellet ($99.75 \pm 0.04\%$ in mode I and $99.45 \pm 0.19\%$ in mode II) were significantly higher ($p < 0.05$) than those in corn straw pellet burning ($98.49 \pm 0.83\%$ in mode I and $97.43 \pm 1.87\%$ in mode II), and high MCE often resulted in low pollutant emissions (Dhammapala et al., 2006; Shen et al., 2010). As a result, differences in EFs between these two biomass pellets can not be explained simply by one or two factors. The impacts of fuel property and combustion condition were complicated and interacted with each other (Johansson et al., 2004; McDonald et al., 2000; Roden et al., 2006).

The profiles of 4 oPAHs were similar between the two types of pellets and between the two modes (S5). 9FO and ATQ were the dominant oPAHs identified, which contributed 26.9 ± 19.5 and $41.5 \pm 9.0\%$ of the 4 total oPAHs from the corn straw pellet burning, and 36.4 ± 19.8 and $45.6 \pm 12.9\%$ of the total from the pine wood pellet burning, respectively.

3.2 Comparison with Emissions from Raw Fuel Burning

Emissions of most pollutants from pellets are expected to be lower than those from raw biomass fuels. In this study, biomass pellets had relatively high ash content, combustion temperature, and combustion efficiency, but relatively low VM content, and moisture in comparison with raw fuels. These differences were likely responsible for the distinct combustion performance and emissions. MCEs for raw corn straw and pine wood combustions in the traditional stove were 96.1 ± 0.4 and $94.1 \pm 1.6\%$, respectively, lower than those of 98.0 ± 1.5 and $99.6 \pm 0.2\%$ for the pellet burning in the modern burner. The measured EFs of oPAHs for the biomass pellets were lower than those for raw biomass, especially in mode I (Figure S4). For example, EF_{oPAHs} for pine wood were 371 ± 266 $\mu\text{g/kg}$, while EF_{oPAHs} for pine pellets were 77.7 ± 49.4 and 348 ± 305 $\mu\text{g/kg}$ in modes I ($p < 0.05$) and II ($p > 0.05$). The insignificant difference in mode II was likely due to the relative large variation in EF measurements. It should be noted that the difference in emission is a result of both shifting in fuel properties (raw to pelletized fuels) and combustion facilities (a brick wok stove to a small modern burner). In addition to MCE, other factors including fuel density, moisture, ash content, combustion temperature and air supply amount could also result in different emission performance between pellet burning and raw fuel combustion. Moreover, the impacts of these factors were often complicated and sometimes interacted with one another. Future study on the influences of specific factors under control conditions is interesting.

3.3 Implication of Pellet Stove Deployment

Reduction of pollutant emission from primary combustion of biomass under wide deployment of pellets can be expected since EFs for the pellets are often lower than those for the raw biomass (Olsson et al., 2003; Johansson, et al., 2004; Boman et al., 2011). Further emission reduction can be realized by increasing thermal efficiencies using modern burners instead of traditional cooking stoves (Chen et al., 2005). It was reported that the thermal efficiency (the ratio of energy absorbed by the water to the energy of consumed fuels) of the pellet burning in a modern burner was about 4~5 times higher than that of raw biomass burning in a traditional cooking stove (Chen et al., 2005).

The ratio between EF_{oPAHs} for pellets burned in the modern burner (EFs in mode I and II collectively applied) and EF_{oPAHs} for the corresponding raw fuels burned in the traditional cooking stove (R_{EF}) was calculated based on the measured EFs. Median R_{EF} values for corn straw and pine wood were 0.369 (0.126–0.541 as inter-quartile range) and 0.378 (0.178–0.835), respectively. By assuming that the thermal efficiency of a modern burners was 4 times of that of a traditional stove (Chen et al., 2005), the emissions would be only 0.092 and 0.095 times of those without deployment. In another word, there would be approximately 90% reduction (S7). Even if there was only 40% of raw fuel combustion in the traditional stoves were replaced with pellets burned in modern burners, the total reduction could be as high as 36%. In addition to replace raw biomass burned in traditional stoves, application of pellets burned in modern burners can also replace coal consumption in residential sector, which can reduce emissions of many hazardous pollutants (Zhang and Smith, 2007) and net emission of CO_2 (Houghton, 2007). Of course, this is a rough estimation based on the limited data available. More field data are needed for providing more accurate results to policy makers.

4. Conclusion

For the first time, EFs of oPAHs from biomass pellet burning in a small modern burner were measured. For the pellets made from pine wood, EF_{oPAHs} were 77.7 ± 49.4 and 348 ± 305 $\mu g/kg$ in combustion mode I (without secondary side air supply) and mode II (with secondary air supply), respectively. For the pellets made from corn straw, EF_{oPAHs} were 189 ± 118 and 396 ± 387 $\mu g/kg$ in mode I and II, respectively. In both two types of pellets and two combustion modes, large variations were found due to differences in fuel properties and combustion conditions. Future studies under controlled conditions to address the impacts of fuel moisture, ash content, combustion temperature, and air supply, are recommended. More field measurements are urgently required.

In comparison with emissions from raw biomass burning in a traditional stove, biomass pellets burned in a modern burner generally had lower emissions. Together with the increased thermal efficiencies of the modern burners, total emissions of oPAHs, as well as other pollutants, from biomass fuel combustion in traditional cooking stoves can be reduced substantially by the deployment of biomass pellets and modern burners. Of course, it should be point out though the emission reduction could be concluded, the quantitative percents based on this study were not representative. The results can not be generalized simply since

data was limited so far. The real benefits of pellet deployment need more data on both emission performance and ambient air pollution.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Table 1

EFs of CO (g kg^{-1}) and 4 oPAHs (mg kg^{-1} , dry basis) for pine wood and corn straw pellets combusted in the modern burner in two different modes without (I) and with (II) secondary side air supply. Mass percent of particulate phase oPAHs to the total (P) was also calculated and listed for each individual. Data shown are means and standard deviations from triplicate experiments.

Fuel	Pine wood pellet		Corn straw pellet	
Burning mode	Mode I	Mode II	Mode I	Mode II
CO	2.69 ± 0.43	6.07 ± 2.04	15.2 ± 8.3	27.1 ± 20.6
9FO	$3.53 \pm 3.46 \times 10^1$	$1.67 \pm 2.15 \times 10^2$	$3.44 \pm 2.33 \times 10^1$	$2.09 \pm 2.80 \times 10^2$
p,%	32.3 ± 17.0	65.0 ± 35.6	56.7 ± 35.6	73.3 ± 19.5
ATQ	$3.19 \pm 1.37 \times 10^1$	$1.36 \pm 0.98 \times 10^2$	$7.69 \pm 4.54 \times 10^1$	$1.36 \pm 1.17 \times 10^2$
p,%	63.8 ± 18.6	68.5 ± 18.6	65.8 ± 27.7	77.8 ± 6.9
BZO	$8.68 \pm 8.03 \times 10^0$	$3.57 \pm 2.52 \times 10^1$	$6.80 \pm 4.93 \times 10^1$	$4.40 \pm 4.86 \times 10^1$
p,%	79.4 ± 33.7	99.7 ± 0.4	68.3 ± 35.5	84.3 ± 22.1
BaAQ	$1.86 \pm 0.91 \times 10^0$	$9.24 \pm 4.01 \times 10^0$	$9.50 \pm 5.04 \times 10^0$	$6.80 \pm 4.56 \times 10^0$
p,%	76.3 ± 17.4	90.4 ± 14.1	85.1 ± 21.3	84.2 ± 13.1
Total OPAHs	$7.77 \pm 4.94 \times 10^1$	$3.48 \pm 3.05 \times 10^2$	$1.89 \pm 1.18 \times 10^2$	$3.96 \pm 3.87 \times 10^2$