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Reductions in Emissions of Carbonaceous Particulate Matter and Polycyclic Aromatic Hydrocarbons from Combustion of Biomass Pellets in Comparison with Raw Fuel Burning

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Abstract

Biomass pellets are emerging as a cleaner alternative to traditional biomass fuels. The potential benefits of using biomass pellets include improving energy utilization efficiency and reducing emissions of air pollutants. To assess the environmental, climate, and health significance of replacing traditional fuels with biomass pellets, it is critical to measure the emission factors (EFs) of various pollutants from pellet burning. However, only a few field measurements have been conducted on the emissions of carbon monoxide (CO), particulate matter (PM), and polycyclic aromatic hydrocarbons (PAHs) from the combustion of pellets. In this study, pine wood and corn straw pellets were burned in a pellet burner (2.6 kW) and the EFs of CO, organic carbon, elemental carbon, PM, and PAHs (EF_{CO} , EF_{OC} , EF_{EC} , EF_{PM} , and EF_{PAH}) were determined. The average EF_{CO} , EF_{OC} , EF_{EC} , and EF_{PM} were 1520 ± 1170 , 8.68 ± 11.4 , 11.2 ± 8.7 , and 188 ± 87 mg/MJ for corn straw pellets, and 266 ± 137 , 5.74 ± 7.17 , 2.02 ± 1.57 , and 71.0 ± 54.0 mg/MJ for pine wood pellets, respectively. Total carbonaceous carbon constituted 8 to 14% of the PM mass emitted. The measured values of EF_{PAH} for the two pellets were 1.02 ± 0.64 and 0.506 ± 0.360 mg/MJ, respectively. The secondary side air supply in the pellet burner did not change the EFs of most pollutants significantly ($p > 0.05$). The only exceptions were EF_{OC} and EF_{PM} for pine wood pellets because of reduced combustion temperatures with the increased air supply. In comparison with EFs for the raw pine wood and corn straw, EF_{CO} , EF_{OC} , EF_{EC} , and EF_{PM} for pellets were significantly lower than those for raw fuels ($p < 0.05$). However, the differences in EF_{PAH} were not significant ($p > 0.05$). Based on the measured EFs and thermal efficiencies, it was estimated that 95, 98, 98, 88, and 71% reductions in the total emissions of CO, OC, EC, PM, and PAHs could be achieved by replacing the raw biomass fuels combusted in traditional cooking stoves with pellets burned in modern pellet burners.

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Note: the authors declare no competing financial interest.

Supporting Information Available The following materials: the photos of the biomass pellets and the pellet burner, size distribution of emitted PM, measured EF_{PAH} , composition profiles and calculated isomer ratios of PAHs for the burning of both raw biomass fuels and pellets, are provided in the Supporting Information and available free of charge via the Internet at <http://pubs.acs.org>.

Introduction

Biomass fuels, mainly wood and crop residues, are important primary energy sources and contribute approximately 13% of the total final fuel consumption worldwide^{1–2}. It has been estimated that 39% of the global population use biomass fuels for cooking and heating and large amounts of biomass fuels are consumed in developing countries³. Without substantial socioeconomic development, the use of biomass fuels in rural areas of developing countries is not expected to change significantly in the future^{3–4}. The burning of biomass fuels is one of the most important sources of air pollutants, including emissions of CO, volatile organic carbons, particulate matter (PM), black carbon, and polycyclic aromatic hydrocarbons (PAHs)^{4–6}. These emissions not only cause severe indoor air pollution and premature death of rural residents^{4–5}, but also contribute to regional and even global air pollution and climate forcing^{7–10}. It has been reported that 1.97 million death (3.3% of the global total) and 41 million disability-adjusted life year (2.7% of the global total) with a global life expectancy of 71.4 years, were attributable to exposure to indoor smoke from solid fuel combustion (biomass fuel and coal collectively) in 2004^{10–11}.

Replacing traditional fuels with cleaner and more affordable fuels is a challenge in most developing countries^{2,12}. Among the various options, biomass pellets, especially those made of crop residues, have great potential because a large volume of crop residues is available in rural areas and would be burned indoors or in the field anyway². The use of biomass pellets has been heavily promoted over the last several years because they are beneficial both from an environmental and social-economic standpoint^{2,13–14}. In Sweden, for example, the annual production of wood pellets was more than 1 million tons in 2005, half of which were consumed in individual households¹⁴, and residential pellet consumption increased steadily at a rate of 30% annually¹³. In China, promotion of biomass pellets has been included in the National Medium- and Long-Term Strategy Plan for Renewable Energy Development and the target goal is 50 million tons of annual consumption by 2020¹².

The pollutant emissions from biomass pellet burning in modern burners were expected to be lower than those from raw fuel combustion in traditional stoves^{15–17} and studies on the emission factors (EFs) of several gaseous pollutants and PM from the burning of pellets have been conducted^{13–18}. EFs often vary dramatically depending on fuel type, combustion facility, and even test method. Reliable and representative EFs are necessary for inventory development. Lacking of emission data is one of the most important reasons causing high uncertainties in current inventories^{19,20}. Therefore, a large number of measurements are needed before meaningful EFs can be obtained. This is especially true for developing countries where the measurements on emission from residential combustions, especially biomass pellet burning, are rather limited.

The objectives of this study were 1) to measure EFs of CO, PM, organic carbon (OC), elemental carbon (EC), and PAHs from the burning of two types of pellets in a commercial pellet burner; 2) to characterize the size distribution of PM and PAH composition profiles from the emission; 3) to compare the emissions from pellet burning and raw fuel combustion in a traditional cooking stove; and 4) to discuss the potential impact of biomass pellet deployment on the total emissions of these pollutants from residential sector.

Method

Fuels and Stove

Two commercial biomass pellets (8 mm in diameter and 1.5–2.0 cm in length) made of pine wood and corn straw, respectively, and a pellet burner (Figure S1) were purchased from a local market in rural Beijing. These pellets and the burner are currently the most popular in

the market. For comparison, raw pine wood (*Pinus tabulaeformis* Carr.) and corn straw (*Zea mays*) were also collected and burned. Fuel properties, including density, moisture, contents of C, H, N, O, volatile matter (VM), ash content, and lower heating value (LHV) were measured and are listed in Table 1. The bulk densities of the pellets were much higher than those of the raw materials, especially for corn straw. The moisture of the two pellets were 5.63 and 5.80%, respectively, and generally lower than those of raw pine wood (9.10%) and corn straw (7.02%). Biomass pellets had lower VM and higher ash contents than the corresponding raw fuels. Ash content of the corn straw pellet was higher than that of pine wood pellet. Similar difference was reported in the literature^{15,21–23}. It was found that ash contents of our pellet fuels were much higher than those made and used in Europe and United States^{15–16,21,24}. A comparison study on properties of biomass pellets also indicated that ash contents of the pellets from China (7.71–21.7 and 1.01–9.25% for crop straw pellets and wood pellets, respectively) were significantly higher than those from Sweden²². It was suggested that these high ash content pellets usually had lower softening temperature and were much easier to form slag. Hence, use of anti-slagging additives was recommended^{22,25}. The slagging rates in the burning of corn straw pellets with 3% of MgCO₃, CaCO₃, Al₂O₃, and kaolin added were reported to be 1.12, 5.07, 25.8, and 43.9%, in comparison with a slagging rate of 46.5% without additives²⁶. The use of additive can reduce slag formation effectively, but increase ash content as well.

Combustion Experiments

The combustion experiments were conducted in a rural kitchen, where previous studies on emissions from indoor crop residue and wood combustions were conducted^{27,28}. For the pellet burning, pre-weighed pellets were added into the burner and fired. After ignition, the burner was set up under a stainless steel hood. The pellets were combusted in the burner in two modes, without (mode I) and with (mode II) secondary side air supply (see Figure S1). For raw corn straw and pine wood, the combustion experiments followed the same procedure used in a previous study²⁷. The same mass of raw corn straw and pine wood as the pellets was burned in a traditional cooking stove in the same kitchen. The smoke from the cooking stove (through a heated bed) and from the pellet burner (through a stainless steel hood and pipe) entered a mixing chamber (4.5 m³) where sampling and on-line measurements were conducted. The combustion experiments were conducted in triplicate.

During the pellet combustion, the kitchen room air was much less smoky in comparison to the room air when the raw materials were combusted in the traditional stove. The chamber temperatures in the pellet burner without the secondary air supply were slightly higher (mode I, 650–900 and 500–800°C for pine wood and corn straw pellets, respectively) than those with secondary air supply (mode II, 550–750 and 400–700°C for pine wood and corn straw pellets, respectively). However, both of these modes had higher temperatures than the traditional cooking stove during the combustions of raw pine wood (350–650°C) and corn straw (300–500°C).

Sampling, Extraction, and Analysis

The procedures of sample collection, extraction, cleanup, and analysis have been previously reported²⁹. Polyurethane foam plugs (PUF, 22 mm diameter × 7.6 cm, 0.024 g/cm³) and quartz fiber filters (QFFs, 25 mm in diameter) were used to collect gaseous and particulate phase air samples, respectively.

Gaseous CO₂ and CO concentrations were measured using an on-line detector equipped with non-dispersive infrared sensor (GXH-3051, Tech. Inst., China) every 2 seconds. The equipment was calibrated using a span gas before each experiment (CO, 1.00%; CO₂, 5.00%). PM mass was measured by weighing the filters before and after sampling using a

0.00001 g digital balance. EC and OC were analyzed using a Sunset EC/OC analyzer (Sunset, USA).

The PUFs were extracted using Soxhlet extraction with 150 ml of dichloromethane for 8 h. QFFs were extracted using a microwave accelerated system (CEM Mars Xpress, USA, 1200 W) with 25 ml of hexane/acetone mixture (1:1, v/v). The temperature program was to 110 °C in 10 min and held for another 10 min. The extract was concentrated to 1 ml and then transferred to a silica/alumina gel column for cleanup (12 cm silica gel, 12 cm alumina, and 1 cm anhydrous sodium sulfate from bottom to top; and pre-eluted with 20 ml hexane). The column was eluted with 70 ml hexane/dichloromethane (1:1, v/v), and the eluate was concentrated to 1 ml and spiked with internal standards for measurement. A gas chromatograph (GC, Agilent 6890) coupled with a mass spectrometer (MS, Agilent 5973) and equipped with a HP-5MS capillary column (30 m × 0.25 mm × 0.25 μm) was used to detect PAHs. The GC oven temperature program was 50°C for 1 min, increased to 150°C at a rate of 10 °C/min, to 240°C at 3°C/min, and to 280°C and held for 20 min. Helium was used as the carrier gas. PAHs were identified based on the retention times and qualifying ions of standards in selected ion monitoring mode. The twenty-eight parent PAHs measured in this study included naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), retene (RET), benzo[c]phenanthrene (BcP), cyclopenta[c,d]pyrene (CcdP), benzo(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), benzo(e)pyrene (BeP), perylene (Per), dibenz(a,h)anthracene (DahA), indeno(1,2,3-cd)pyrene (IcdP), benzo(g,h,i)perylene (BgHiP), dibenzo[a,c]pyrene (DacP), dibenzo[a,l]pyrene (DalP), dibenzo[a,e]fluoranthene (DaeF), Coronene (Cor), dibenzo[a,e]pyrene (DaeP), dibenzo[a,i]pyrene (DaiP), and dibenzo[a,h]pyrene (DahP). Deuterated PAHs (Nap-*d*₈, Ant-*d*₁₀, Ane-*d*₁₀, Chr-*d*₁₂, and Perylene-*d*₁₂, J&W Chemical Ltd., USA) were used as internal standards.

Quality Control and Data Analysis

The preparation of PUFs and QFFs for sampling has been previously described²⁹. In brief, the GFFs were baked at 450°C for 6 h and stored in a desiccator. The PUF was pre-extracted using acetone, dichloromethane, and hexane in sequence for a total of 24 h. After sampling, the PUFs and QFFs were packed in aluminum foil. All glassware was cleaned using an ultrasonic cleaner and baked at 500°C for 10 h. Procedural blanks were measured and subtracted from the sample measurements. For PAHs, the instrumental detection limits ranged from 0.13 ng (ACY) to 0.92 ng (BgHiP). Method detection limits ranged from 0.23 ng/mL (NAP) to 1.42 ng/mL (BgHiP) for gaseous PAHs and from 0.53 ng/mL (PHE) to 1.32 ng/mL (BgHiP) for particulate phase PAHs. Recoveries of the spiked standards ranged from 70 to 121% for gaseous phase PAHs, and 68 to 120% for particulate phase PAHs.

The EFs were calculated using the carbon mass balance method with the assumption that the carbon in the fuel was released in the form of CO₂, CO, total gaseous hydrocarbons, and carbonaceous carbon in the particulate matter. EFs are given in energy units (mg/MJ) using LHV_s of burned fuels for unit conversion¹⁹. Modified combustion efficiencies (MCE) were calculated as CO₂/(CO₂+CO) ratios (molar basis). For the purpose of comparison, a relative difference (RD) was calculated as (EF_{pellet} - EF_{raw})/EF_{raw} where EF_{pellet} and EF_{raw} are EFs of a given pollutant for biomass pellets (measured EFs in mode I and mode II collectively applied) and the corresponding raw fuel, respectively. The potential reduction in total emissions of CO, OC, EC, PM, and PAHs were also estimated based on the differences in the calculated EFs between the pellets and raw fuels, reported thermal efficiencies of cooking stoves and modern burners, and emission inventories in the literature. The uncertainties of the RDs and the total emission reductions were estimated using Monte Carlo simulations, which were run 100,000 times with coefficients of deviation of 0.10 for total

emission, 0.20 for thermal efficiency, and the calculated means and standard derivations for the measured EFs. Statistica (v5.5, StatSoft) was used for statistical analysis with a significance level of 0.05.

Results and Discussion

EFs of OC, EC, and PM for Biomass Pellets

The measured EFs of CO, OC, EC, and PM (EF_{CO} , EF_{OC} , EF_{EC} , and EF_{PM}) for the two types of biomass pellets (pine wood and corn straw) in the two different combustion modes (with and without secondary side air admission) are listed in Table 2 as means and standard derivations. In general, EFs for corn straw pellets were higher than those for pine wood pellets ($p < 0.05$). The measured values varied significantly, even among the triplicate combustion experiments for the same fuel in the same combustion mode, indicating high variability in the emission. For example, the coefficients of variation for EF_{OC} , EF_{EC} , and EF_{PM} for pine wood pellets were 78, 84, and 27% in mode I (without secondary side air admission) and 84, 82, and 42% in mode II (with secondary side air admission), respectively.

As expected, EF_{OC} and EF_{PM} for pine wood pellet combustion in mode II were significantly higher than those in mode I ($p < 0.05$), because the combustion temperature was cooled down by the extra air supply^{15–16}. EF_{EC} for the pine wood pellets and EF_{OC} , EF_{EC} , and EF_{PM} for corn straw pellets were not significantly different between the two modes ($p > 0.05$), though the average EF_s measured in mode II were higher than those in mode I. Similarities in EF_s of PCDD/F, PCB_s, and HCB for firewood and pellet combustions between full and reduced air supplies have been previously reported³⁰.

The OC/TC ratio ($TC=OC+EC$) and total carbon mass fraction in PM (TC/PM) are useful indicators in source apportionment and emission inventory development for carbonaceous PM^{19,31–32}. The calculated OC/TC and TC/PM were 0.40 ± 0.20 and 14% for corn straw pellets, and 0.61 ± 0.29 and 8.2% for pine wood pellets, respectively. There was no significant difference in these ratios between the two combustion modes ($p > 0.05$) and the OC/TC ratio was similar to that for *Miscanthus* and *Triticale* pellets reported in the literature (0.52 ± 0.26)²¹.

EF_{CO} and EF_{PM} for pellets reported in the literature varied dramatically among various fuel types and burning conditions^{15,16,18,21,33–35}. For example, for *Triticale* pellets, *Miscanthus* pellets, and wood pellets burned in a 40 kW burner, EF_{PM} were 114, 14.7, and 21.9 mg/MJ (average for start-up, full load, and part load phases), respectively²¹. For wood pellets burned in two pellet burners under different burning conditions (3–22 kW), the measured EF_{CO} and EF_{PM} were in the range of 30–1100 and 12–65 mg/MJ¹⁵. In a stove with output energy of 3–5 kW (very close to 2.6 kW in our study), EF_{CO} and EF_{PM} for wood pellets were measured at 57–270 and 19–58 mg/MJ¹⁸. Boman *et al.* summarized the published EFs for pellets in the literature and reported that EF_{CO} and EF_{PM} were 58–2600 and 11–600 mg/MJ in the field measurements, and 24–2000 and 2–150 mg/MJ in the laboratory studies, respectively¹⁶.

By thoroughly reviewing reported values in the literature, it was summarized that average EF_{PM} for wood pellets in residential wood combustion were 1.91 and 3.99 g/kg in U.S. EPA certified (1988 New Source Performance Standards for woodstoves) and exempt modes, respectively³⁶. For pellet heaters used in the MANE-VU region (11 states and the District of Columbia), mean EF_{PM} was 1.53 g/kg based on measurements in old, modern, EPA-certified, and EPA-exempt models and pellet heating systems¹⁷. It was also recognized that for wood pellets burned under high efficiencies, EF_{PM} was about 10–50 mg/MJ^{37–38}. Our

results for EF_{CO} were 136–505 mg/MJ for pine wood pellets and 414–3838 mg/MJ for corn straw pellets. EF_{PM} were 17.6–176 and 65.5–332 mg/MJ for these two pellets, respectively. Average EF_{PM} for pine wood pellets was 71.0 ± 54.0 mg/MJ (1.17 ± 0.89 g/kg). Generally, the results fell within the reported range in the literature.

Measured EF_{OC} and EF_{EC} for biomass pellets were rather limited. In a 25 kW burner, EF_{EC} and EF_{OC} for wood pellets were reported at 0.1 ± 0.17 and 0.9 ± 2.1 mg/MJ³⁴. In an inventory on carbonaceous PM emission in Europe, 0.83 mg/MJ was adopted for both EF_{EC} and EF_{OC} for biomass burned in an automatic feed pellet boiler (< 50 MWh) under uncontrolled condition³⁹. These values were lower than the EF_{OC} (5.74 ± 7.17 , 0.335–4.60 mg/MJ) and EF_{EC} (2.02 ± 1.57 , 0.223–18.5 mg/MJ) for pine wood pellets measured in our study (results from both two modes). The difference could be due to the different pellet burners and fuel types. The exact reason for such difference, as well as the difference on the estimated total emission and the impacts on air quality and climate forcing, should be further investigated.

The PM size distributions of the pellet combustion emissions were characterized using a nine-stage cascade impactor with cut-off diameters of 0.4, 0.4–0.7, 0.7–1.1, 1.1–2.1, 2.1–3.3, 3.3–4.7, 4.7–5.8, 5.8–9.0, and 9.0–10.0 μm . As shown in Figure S2, PM size distributions were not significantly different between the two combustion modes. PMs with diameter less than 0.4 μm ($PM_{0.4}$) were the most abundant, contributing 31.6 ± 9.0 and $20.3 \pm 1.9\%$ of the total PM_{10} from corn straw and pine wood pellet combustions, respectively. $PM_{1.1}$ (most close to $PM_{1.0}$) and $PM_{2.1}$ (most close to $PM_{2.5}$) made up 54.5 ± 5.9 and $63.4 \pm 7.1\%$ of the total PM_{10} for pine wood pellets and 70.5 ± 6.1 and $79.9 \pm 4.5\%$ of the total PM_{10} for corn straw pellets, respectively. Calculated mass media diameters (MMDs) for corn straw pellets were 0.62 ± 0.24 (0.29–0.92) and 1.2 ± 0.2 (1.0–1.5) μm for pine wood pellets.

The prevalence of fine PM in pellet burning emission has been reported in the literature. By using a Dekati Low-Pressure Impactor (DLPI), Johansson *et al.* found that size distributions of particles from two pellet burners were similar with a peak size of 130 nm¹⁵. Båfver *et al.* reported a high mass fraction of $PM_{1.0}$ (81–94%) and $PM_{2.5}$ (84–96%) in particles from wood pellet burning in several pellet stoves of 3–5 kW output power¹⁸. Also using a DLPI, Lamberg *et al.* measured size distribution of PM for wood pellets combusted in a 25 kW burner, and calculated a MMD value of 137 ± 6.1 nm³⁴. In general, $PM_{1.0}$ contributed approximately 90% of the total PM emitted in pellet burning⁴⁰. However, the concentration and distribution can be influenced by fuel property (e.g. moisture and ash content) and combustion conditions (e.g. air supply and combustion temperature)^{15,16,40}. It was found that pellet burning can also produce relative large amounts of coarse PM under given conditions^{16,23}. For example, as high as 15% $PM_{1.0-10}$ was found in PM emissions from wood pellet burning in a 5–6 kW stove. And in the startup phase in a 2 kW burner under intermittent operation, $PM_{1.0}$ only made up 70% of the total¹⁶.

In comparison, pellet burning in our study produced more coarse particles. The difference is likely due to the distinct pellets (fuel property) and burners used (combustion condition)^{15,16,18}. The biomass pellets used in this study generally had higher ash content, which could be associated with the size distribution of PM released^{16,23}. Although there is no direct evidence on the cause of such high ash content, additives used in the pellet manufacture can not be ruled out. In fact, addition of magnesium and calcium carbonates, aluminum hydroxide, kaolin and so on as additives was often recommended for pellet manufacturing in China^{22,26}. Smelting slag observed after burning in this study (Figure S3) was an indirect evidence. Yao *et al.* found that silicon contents in the ash from pellets made in China were 10 to 25% higher than those made in Sweden²². Such a difference could not be only explained by the difference in the composition of wood or crop residue.

Unfortunately, chemical compositions of the pellets or ash were not measured in this study. Therefore, future studies focusing on the impacts of various additives on the fuel properties (e.g. volatile matter, ash content, and heating value) and emission characterizations (e.g. emission factors, chemical compositions and size distributions of PM and PM-bound pollutants) are strongly recommended. Also, information on mineral compositions of the fuel and emitted particle, and their impacts on PM size distribution are required in the future to address such difference.

EFs of PAHs for Pellets

EFs for the sum of the 28 PAHs measured (EF_{PAH}) for pine wood and corn straw pellet combustions were 0.329 ± 0.261 and 0.711 ± 0.325 mg/MJ in mode I, and 0.718 ± 0.369 and 1.32 ± 0.76 mg/MJ in mode II, respectively. There was no statistical difference between these two types of pellets ($p > 0.05$). The sixteen U.S. EPA priority pollutant PAHs contributed more than 90% of the total PAHs measured. No significant difference was identified between the two combustion modes ($p > 0.05$), although the measured mean EFs in mode II were always higher than those in mode I (Table S1). It is possible that the extra air supply enhanced the formation of incomplete combustion products by reducing temperature^{15–16}. In addition, the large variation in EF_{PAH} among the triplicate experiments resulted in no significant difference.

There were several reports on PAH emissions from wood pellet combustion^{15–17,36}. EFs of 16 PAHs and BaP for pellet heaters in the MANE-VU region were estimated at 350 and 3.34 mg/kg, respectively¹⁷. For wood pellets burned in residential burners operated under 3–22 kW, EFs of 27 particle-bound PAHs ranged from 0.06 to 8.5 mg/MJ¹⁵. In another study, the total EFs of 40 PAHs in both gaseous and particulate phases were only 0.002–0.34 mg/MJ¹⁶. Our results (EFs of 28 PAHs were 0.0942–1.16 mg/MJ, with 0.0091–0.660 mg/MJ for PM-bound PAHs) also varied widely with no significant difference with the reported values. Taking strong health impact of PAHs, large contribution of PAHs from biomass burning sources, and potential benefit of replacing the traditional biomass fuels with biomass pellet fuels in emission reduction into consideration^{16,41}, more investigations on emissions and factors affecting the emissions are urgently needed.

On average, the total PAHs constituted only 0.20% (0.040 to 4.0%, as the range) and 0.14% (0.022 to 0.29%, as the range) of the total mass of the PM from the combustions of pine wood and corn straw pellets, respectively, comparable to 0.10% (0.002 to 0.45%, as the range) reported in the literature¹⁶. The PAH/PM ratios in the two combustion modes were not statistically different ($p > 0.05$). Low and median molecular weight PAHs dominated the total PAHs emitted in both corn straw and pine wood pellet burning (Figure S4). NAP and PHE were the most abundant, similar to those reported in the literature¹⁶. The PAH profiles were not significantly different between two modes ($p > 0.05$). To provide baseline information for PAH source apportionment, 6 commonly used isomer ratios, including ANT/(ANT+PHE), FLA/(FLA+PYR), BaA/(BaA+CHR), IcdP/(IcdP+BghiP), BbF/(BbF+BkF), and BaP/(BaP+BghiP) were calculated and listed in Table S2. There were no significant differences in these ratios between the pine wood and corn straw pellets or between combustion mode I and II ($p > 0.05$).

Comparison between the Biomass Pellets and Raw Fuels

To compare the EFs of OC, EC, PM, and PAHs for biomass pellets and raw fuels, these EFs were also measured from the combustion of pine wood logs and corn straw in a traditional cooking stove. The results are listed in Table 2 (CO, OC, EC and PM) and Table S1 (PAHs) as well. In comparison with the raw materials, biomass pellets had higher bulk density and ash content, but lower VM content. During the experiments, the kitchen was much less

smoky when pellets were burned in the modern burner as compared to the raw fuel being burned in the traditional stove. In general, the measured chamber temperatures in the pellet burner (550–900°C during pine wood pellet combustion and 400–800°C in corn straw pellet burning) were higher than the residential stove (350–650°C during pine wood combustion and 300–500°C in corn straw burning), and the MCEs for the burning of raw pine wood log (94.1%) and corn straw (96.1%) were lower than those for pellet combustion (Table 2). For the same mass, pellet burning usually took longer than raw material burning. As a result, the calculated burning rates for pellets were lower than for raw fuels (Table 2).

Significant reduction ($p < 0.05$) in EF_{CO} , EF_{OC} , EF_{EC} , and EF_{PM} for both pine wood and corn straw pellets were found in comparison with EFs for the raw fuels, except EF_{PM} of 112 ± 48 mg/MJ for pine wood pellet in mode II that was not significantly different from, though looked slightly higher than, EF_{PM} for raw pine wood at 95.3 ± 19.2 mg/MJ ($p > 0.05$). The calculated RD values (see **Method** section) for CO, OC, EC, and PM were -55 (-71 to -31 , as inter-quartile range from Monte Carlo simulation), -90 (-91 to -88), -68 (-77 to -54), and -49% (-63 to -30%) for corn straw, and -89 (-91 to -88), -93 (-96 to -86), -97 (-98 to -56), and -43% (-60 to -18%) for pine wood, respectively. RDs for OC and EC were much lower than those for PM, suggesting that pelleting can reduce the emissions of OC and EC more effectively than PM. As a result, TC/PM ratios for pine wood (8%) and corn straw (14%) pellets were significantly lower than those for raw pine wood log (97%) and corn straw (28%) ($p < 0.05$). Relatively low organic fractions, but high inorganic fractions, in PM from pellet combustions have been reported in the literature^{16,18}.

Figure 1 shows the cumulative size distributions of PM from the combustion of raw and pelletized corn straw (A) and pine wood (B). Higher percents of coarse PM were in the smoke emitted from pellet burning in comparison with those for raw fuels. The size distribution of PM can be affected by many factors including fuel moisture, ash content, combustion temperature, excess air, and combustion efficiency^{23,40,42}. Moreover, the impacts of these factors were complicated and sometimes interacted with one another. PM size is very sensitive to the combustion temperature that varies due to different fuel moisture and excess air^{42,48,49}. Combustion efficiency could be influenced by various factors, like fuel moisture, mass load, and excess air^{46,47}. It was found that geometric mean diameters of PM were lower for residential wood combustion under higher combustion efficiency^{43–44}, but a positive linear relationship between median diameter and MCE was reported for fresh forest smoke⁴⁵.

In comparison with raw fuels, biomass pellets in this study had relatively high ash content, combustion temperature, and combustion efficiency, but relatively low VM content and moisture. These differences are likely responsible for the distinct size distributions. Biomass pellets are usually made through processes of comminuting, drying, pelletizing and cooling. Several key parameters in pellet manufacturing are fuel moisture, grind size, applied pressure, and specific energy requirement⁵⁰. To avoid potential disintegration, raw material can be conditioned sometimes, by adding binding agents or steam application⁵¹. For the former, some binding agents, such as starch, natural paraffin, molasses, lignin sulfate, and kaolin could be used under the permit of legislation^{51,52}. It was thought that some binders, like kaolin, calcium and magnesium oxide can also hinder the slag formation during combustion^{51,53}. Addition of additives is regulated by some^{24,52}, but not most countries (e.g. CEN/TS 14588: 2004 published by the European Standard Committee ECN/TC 335)²⁴. In China, such standard for pellet quality does not even exist.

Unlike the practice in Europe and North America where additives are usually not used in pellet production, various additives were recommended for preventing slag formation in the biomass pellet manufacturing in China^{25–26}. Although the detailed information on additives

as binding agents or ash inhibitory in the tested pellets were not provided by the manufacturer, the use of additives is likely according to the observed melting slag formation in the bottom of the burner after combustion (Figure S3). In future study, composition and quantity of additives in these pellets and the influences of various additives on the pollutant emissions should be investigated.

Unlike EF_{CO} , EF_{OC} , EF_{EC} , and EF_{PM} , EF_{PAH} for the pellets were higher than for the raw fuels, though the differences were not significant due to high variability ($p > 0.05$). RDs for PAHs were 64% (56 to 72%) for corn straw and 6% (−16 to 33%) for pine wood, respectively. The PAH composition profiles (Figure S4) and calculated isomer ratios (Table S2) were similar between the pelletized and raw fuels. Also, PAH/PM ratios for the pellets were comparable to those for raw fuels ($0.12 \pm 0.02\%$ for pine wood and $0.07 \pm 0.03\%$ for corn straw) ($p > 0.05$).

In the literature, some biomass pellet EF_{PAH} and EFs for other organic pollutants, were not significantly lower than, and sometimes slightly higher than, EFs for raw fuels^{15,30}. For example, unexpected high emissions of PCDD/Fs and PCBs were observed during combustion of wood (intermittent) and straw (continuous) pellets³⁰. It was also reported that higher emissions of toxic organic compounds, including benzene, naphthalene and furan, occurred in the after-flame phase of pellet burning but not in raw oat straw combustion¹⁴. Relatively high ash contents and combustion temperatures are likely responsible for the slightly high EF_{PAH} of the pellet burning compared to that for raw fuels. A better understanding of the effects these different variables have on EF needs further study. In particular, the effect of pellet additives on pollutant emissions needs further investigation⁵⁴.

Implications

Except for EF_{PAH} , the EFs of many air pollutants for pine wood and corn straw pellets are lower than those for raw fuels. The ratios of EF_{pellet} and EF_{raw} (R_{EF}) were calculated for CO, OC, EC, PM, and PAHs based on the measured results. The uncertainty of the calculation was estimated using Monte Carlo simulation. Median R_{EF} (both results of corn straw and pine wood used collectively) of CO, OC, EC, PM, and PAHs were 0.22 (0.13–0.36, as inter-quartile range), 0.090 (0.057–0.14), 0.10 (0.067–0.15), 0.54 (0.40–8.61), and 1.3 (1.0–1.6), respectively. In addition, the thermal efficiency of burning pellets in the modern burners was approximately 2.3 times that of raw fuels burned in the so-called improved cooking stoves and 4.5 times that of traditional cooking stoves⁵⁵. Taking the relatively low EFs (except for the slightly high EF_{PAHs}) and high thermal efficiency of pellet burning in the modern burner into consideration, the replacement of raw fuels with biomass pellets can reduce the emissions of most air pollutants significantly. As a rough estimation, the emission reductions of CO, OC, EC, PM, and PAHs would be 90 (84–94), 96 (94–98), 96 (93–97), 76 (71–80), and 43% (26–56%), respectively, by replacing the improved cooking stoves with pellet stoves. The reduction could be as high as 95 (92–97), 98 (99–79), 98 (96–99), 88 (85–90), and 71% (62–77%) if the traditional cooking stoves were replaced with pellet stoves. Currently, residential biomass burning contributed approximately 39.4, 2.29, 0.59, 3.87, and 0.065 Tg of total emissions of CO, OC, EC, PM, and PAHs in China, respectively^{5,6,56}. If only 20% of the traditional cooking stoves can be replaced with pellet stoves, the total emission reductions in CO, OC, EC, PM, and PAHs would be 7366 (6262–8642), 446 (382–521), 115 (98–134), 675 (577–791), and 8.94 (7.27–10.8) Gg. Of course, this is a very rough estimate based on the result of a single experiment. More studies, especially field studies, are recommended for reducing the uncertainty in this prediction. Even with the uncertainties in this estimation, the environmental benefit could be significant.

Although biomass pellets are undergoing fast deployment in both developed and developing regions, there are rather limited studies to address emissions of various pollutants. In this study, we measured EF_{CO} , EF_{OC} , EF_{EC} , EF_{PM} , and EF_{PAH} for two biomass pellets burned in a commercial pellet burner in two modes. The measured EFs varied half to one order of magnitude. The EFs for the pellets were significantly lower than those for the raw biomass fuels ($p < 0.05$), except EF_{PAH} . Taking both EFs and the stove thermal efficiency into consideration, significant reductions in the total emissions of these pollutants from residential sector are expected by replacing raw fuels combusted in traditional/improved stoves with biomass pellet fuels in modern burners.

It should be noted that only two types of biomass pellets were tested in a small burner with power of 2.6 kW in this study. The results under given conditions can not be simply generalized. It can be concluded that the emission reduction can be achieved by replacing raw fuels with pellets. However, the reduction percentages calculated in this study are not representative. More data, especially field data, should be collected before the reduction can be quantified. Also, factors affecting the emissions, including fuel property, stove type, and combustion condition, should be further investigated.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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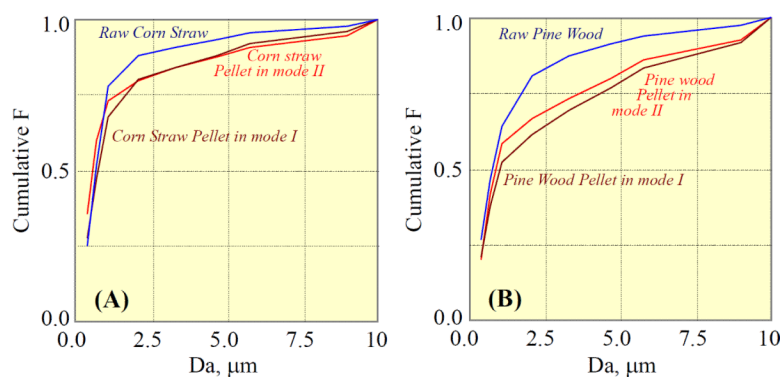


Figure 1. PM size distributions of the pellet and raw fuel burning. (A) Corn straw, (B) Pine wood

Table 1

Fuel properties (dry basis) of raw and pelletized corn straw (*Zea mays*) and pine wood (*Pinus tabulaeformis* Carr.).

	Raw pine wood	Pelletized pine wood	Raw corn straw	Pelletized corn straw
Moisture, %	9.10	5.63	7.02	5.80
Density, g/cm ³	0.44	1.30	0.069	1.41
Proximate Analysis, %				
Ash content	0.25	3.75	4.01	10.84
Volatile matter	84.77	76.77	77.94	70.62
Fixed carbon	14.98	19.48	18.05	18.54
LHV, MJ/kg	16.72	16.48	15.44	13.89
Elemental analysis, %				
N	0.18	0.16	0.96	1.23
C	49.10	46.78	43.47	43.14
H	6.32	5.84	6.02	5.48
O (<i>by difference</i>)	44.41	47.23	49.56	50.16

MCE, burning rate (R), and Efs of CO, OC, EC and PM (mg/MJ) for raw and pellets in this study. Data listed are means and standard derivations from triplicate experiments. For pellets in the burner, two different burn modes were tested, without (I) and with (II) secondary side air admission.

Table 2

Fuel	Raw pine wood		Pine wood Pellet		Raw corn straw		Corn Straw Pellet	
	Mode I		Mode II		Mode I		Mode II	
MCE	0.941±0.016	0.998±0.000	0.994±0.002	0.961±0.004	0.985±0.008	0.974±0.019	1.84±0.36×10 ⁻²	1.95±1.49×10 ³
R, kg/min	7.79±1.03×10 ⁻²	2.12±0.15×10 ⁻²	1.67±0.52×10 ⁻²	4.47±0.20×10 ⁻²	1.52±0.17×10 ⁻²	1.84±0.36×10 ⁻²	1.30±1.55×10 ¹	8.35±1.86×10 ⁰
CO	2.30±0.71×10 ³	1.63±0.26×10 ²	3.68±1.24×10 ²	2.42±0.20×10 ³	1.10±0.60×10 ³	1.30±1.55×10 ¹	1.72±0.94×10 ²	2.04±0.84×10 ²
OC	3.61±2.09×10 ¹	7.00±5.46×10 ⁻¹	9.11±7.64×10 ⁰	5.69±2.37×10 ¹	4.34±0.97×10 ⁰	1.30±1.55×10 ¹	1.30±1.55×10 ¹	1.30±1.55×10 ¹
EC	5.64±2.37×10 ¹	2.07±1.73×10 ⁰	1.99±1.63×10 ⁰	2.60±0.19×10 ¹	1.41±1.20×10 ¹	1.30±1.55×10 ¹	1.30±1.55×10 ¹	1.30±1.55×10 ¹
PM	9.53±1.92×10 ¹	2.96±0.81×10 ¹	1.12±0.48×10 ²	3.01±0.04×10 ²	1.72±0.94×10 ²	1.72±0.94×10 ²	1.72±0.94×10 ²	1.72±0.94×10 ²