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Significant reduction in air pollutant emissions from household cooking stoves by replacing raw solid fuels with their carbonized products

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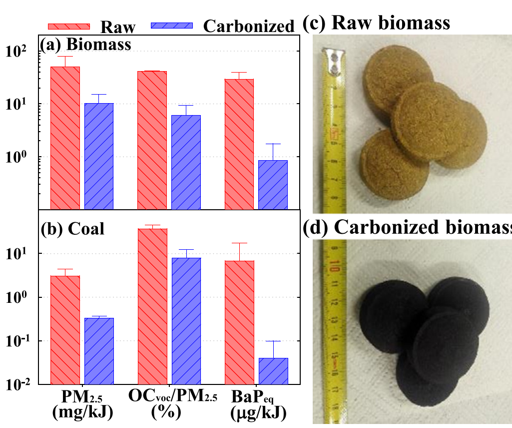
HIGHLIGHTS

- Carbonization treatment of biomass and coal can produce clean solid fuels.
- Carbonized fuels possess higher thermal efficiencies and lower pollutant emissions.
- Reduction of energy delivered-based $PM_{2.5}$, OC/EC and BaP_{eq} are ~80%, 96%/92%, and 95%.
- Switching to carbonized fuels can achieve both environmental and health benefits.
- The reduction is mainly attributed to the removal of fuel's volatile matter content.

GRAPHICAL ABSTRACT



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ABSTRACT

Residential solid fuel combustion contributes significantly to ambient and indoor air pollutions. An appropriate clean solid fuel to reduce residential emissions is urgently needed. This study evaluates the reduction in pollutant emissions achieved by carbonized solid fuels in residential cooking practice. Four biochar samples, three semi-coke briquette samples and their raw materials were tested in a typical cooking stove. These carbonized samples showed higher thermal efficiencies and lower particulate matter (PM) emission factors (EFs) than their raw material samples. Owing to distilled volatile matter during carbonization treatment, average energy delivered-based $PM_{2.5}$ EFs were 10 ± 5 mg/kJ (carbonized) and 50 ± 28 mg/kJ (raw) for the biomass and 0.33 ± 0.04 mg/kJ (carbonized) and 3.0 ± 1.3 mg/kJ (raw) for the coal samples. The energy delivered-based EFs of organic carbon, elemental carbon, and 16 priority polycyclic aromatic hydrocarbons extracted from $PM_{2.5}$ samples from carbonized fuels were reduced by 97 ± 1%, 93 ± 3%, and 97 ± 2%, respectively, for the tested biomass samples, and those for the tested coal samples were 96 ± 1%, 90 ± 6%, and 98 ± 2%, respectively. Average EFs of benzo[*a*]pyrene equivalent carcinogenic potency for individual polycyclic aromatic hydrocarbons were reduced 95 ± 3% to ~0.51 μg/kJ (carbonized) from ~19.6 μg/kJ (raw). Furthermore, the average ratio of volatile organic compounds contained in $PM_{2.5}$ samples was also reduced from 38.8 ± 5.4% to 7.1 ± 3.9%. These results suggest that

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carbonized solid fuels exhibit better performance in reducing carcinogenic potency and pollutants, most of which are highly correlated with the volatile matter content of the fuel. Switching from raw solid fuel to carbonized solid fuel will help to reduce pollutant emissions from household combustion and achieve both environmental benefits and health benefits for household residents.

1. Introduction

Worldwide, approximately 2.7 billion people rely on the tradition use of biomass and 0.4 billion people rely on coal for household combustion, and these numbers are expected to increase in the next two decades (IEA, 2011). For example, 43.4% and 29.5% of China's households utilized these solid fuels for cooking and heating in 2012, respectively (Duan et al., 2014). The combustion of solid fuels in inefficient traditional stoves significantly affects human health via $PM_{2.5}$ (particulate matter with aerodynamic diameter less than or equal to 2.5 μm) emissions (Cai et al., 2018; Lim et al., 2012; Zhang and Smith, 2007), which can also influence climate and visibility (Che et al., 2007; Smith and Bond, 2014). Primary $PM_{2.5}$ from household combustion is mainly composed of organic carbon (OC), elemental carbon (EC), polycyclic aromatic hydrocarbons (PAHs), and toxic elements (Chen et al., 2015a; Li et al., 2017; Zhang et al., 2014; Zhang et al., 2008). Because of incomplete combustion and the lack of air pollution control devices, household stoves have significantly higher emission factors (EFs) than industrial boilers (Chen et al., 2015a; Chen et al., 2006; Chen et al., 2009; Zhang et al., 2014; Zhang et al., 2008; Zhi et al., 2008). Indeed, approximately 33%–46% of primary $PM_{2.5}$, 82–91% of primary OC, 46%–67% of EC, and 62% of PAHs in the atmosphere are emitted by household solid fuel combustion in China (Huang et al., 2014; Lei et al., 2011; Wang et al., 2014; Zhao et al., 2013). Thus, household air pollution has been identified as one of the most important environmental risk factors (Du et al., 2018; Li et al., 2017; Shen, 2015), and caused premature deaths of around 2.8 million in the world and around 1.0 million in China in 2015 (Cohen et al., 2017). $PM_{2.5}$ and PAHs levels in most Chinese rural houses exceed the World Health Organization standard (Du et al., 2018). Other developing countries, such as India and Nepal, have similar problems (Cohen et al., 2017; Sahu et al., 2011).

The adoption of clean fuel to reduce household air pollution has been intensively investigated (Shen, 2015). Volatile matter content and burning form of solid fuels have been wildly recognized as two dominant factors to influence PM emissions and their carbonaceous compositions (Chen et al., 2015a; Li et al., 2016c). The burning of crop straws, wood, and coal chunks in household stoves has high pollutant EFs but low thermal efficiencies, whereas pelletized biofuels and coal briquettes were reported to have lower pollutant EFs (Bond et al., 2002; Chen et al., 2015a; Shen, 2015). However, particle-bound PAHs (the dominant form of PAHs in household flue gas) from coal briquettes were also reported to have significantly higher EFs than those from coal chunks (Chen et al., 2015b), whereas the toxic elemental EFs from pelletized and uncompressed biomasses were identical in terms of the units of energy delivered (energy delivered-based EFs). EFs of PM, EC, and OC from residential solid fuel combustions fast increase with an increase in the solid fuel's volatile matter content, owing to incomplete combustion (Li et al., 2017). PM EFs for anthracite coal were reported to be several ten times lower than that for bituminous coal and raw biomass due to low volatile matter content (Chen et al., 2006; Li et al., 2016c). Thus there are still reduction potentials for household solid fuel combustion, especially the reduction of the fuel's volatile matter content.

Because of the recent development of industrial low-temperature carbonization, semi-coke and biochar powders can be abundantly produced from low-rank coal and biomass in China (Qi et al., 2018; Yang et al., 2016) and other countries (Amarasekara et al., 2017; Mau and Gross, 2018; Minaret and Dutta, 2016; Yeoh et al., 2018), respectively. These raw solid fuels are heated at 450 °C to 700 °C in the absence of air to distill out synthetic fuels, unconventional oil and syngas as clean fuels for commercial energy. Semi-coke and biochar powders have low volatile contents. The current production of semi-coke powders could satisfy half of the household coal demand in China, whereas the industrial carbonization project of biomass in China remains limited to several pilot programs. The major organic species released in the flue gases during the carbonization process are non-aromatic hydrocarbons, carboxylic acids, and aromatic compounds. Thermogravimetric analysis (TGA) and semi-empirical models suggested that the pyrolysis products are the precursors of black carbon and organic aerosols (Brown and Fletcher, 1998; Wang et al., 2015). Thus, the combustion of carbonized fuels may result in lower emission aerosols, whose precursors are released during the industrial carbonization process and collected to make fuel oil and other products. Semi-coke briquettes have recently been reported to exhibit relatively low mass-based PM EFs for household heating activities (Li et al., 2016b,d; Tian et al., 2018). However, the application of these carbonized fuels in households has rarely been evaluated in terms of pollutant emissions and energy efficiencies. Thus, the energy delivered-based EFs of toxic species from typical household cooking activities using these carbonized fuels remain unknown.

The objective of this study is to evaluate the environmental and health benefits of using carbonized coal and biomass briquettes as household cooking fuels in the same burning form. Their raw fuels were also investigated for comparison to measure both the energy delivered-based and mass-based EFs of PM, OC, EC, PAHs, benzo[*a*]pyrene (BaP) equivalent carcinogenic potency (BaP_{eq}) and toxic elements in a traditional cooking stove. The environmental impacts and pollutant-reduction mechanisms are discussed with regarding to the comparison of fuels' volatile matter content and pollutant emission factors.

2. Experimental materials and methods

2.1. Tested solid fuel samples

Four raw biomass samples, four biochar samples, three semi-coke samples, and three raw coal briquettes were tested. To eliminate the effect of fuel burning form, all samples were produced using the same briquetting technique, i.e., power diameters of <1 mm, an adhesion agent in a 5% mass ratio, a molding pressure of 25 MPa, and a briquette diameter of 3 cm. Table 1 presents the quality information describing the samples, including the moisture, ash, volatile matter, fixed carbon, sulfur, nitrogen, carbon, and hydrogen contents and the net calorific value as received. All samples were divided into 7 groups according to their original materials. The four biomass materials, i.e., wheat straw, rice straw, maize straw, and sawdust, were collected from Xuzhou in Jiangsu province, and the three coal samples and their semi-coke powders were collected from Shenmu, Hami, and Xilinguole in Shaanxi, Xingjiang, and Inner Mongolia provinces, respectively. These semi-coke powders were made from their raw coals by industrial carbonization treatment at 500–600 °C. These carbonized biomass samples were produced in the same laboratory carbonization furnace using the identical thermal process with a heating rate of 1 K/min to 500 °C and a subsequent heating at 500 °C for 2 h in the absence of air.

Most volatile matters in coal and biomass were distilled out during the carbonization process as a coke oven gas and sent to the recovery