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Influences of coal size, volatile matter content, and additive on primary particulate matter emissions from household stove combustion

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HIGHLIGHTS

- Incomplete combustion of coal volatile matter leads to significant PM emission from household stove.
- PM emission decrease with increasing coal size and mineral additive in household stove.
- Controlling coal properties helps to reduce PM emission from household coal combustion.

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ABSTRACT

Aiming to reduce primary particulate matter (PM) emission from household coal combustion, we conducted experimental investigations on several coal properties that affect PM emission factors (EFs). Fourteen coal chunk samples with various volatile matter contents on dry and ash-free basis (V_{daf}, 2.84–48.7%) were tested to examine the effect of coal volatile matter content. Eight coal briquette samples with various mineral additives were tested to examine the effect of coal additive. Two coal chunk samples with V_{daf} of 32.9% and 9.3%, respectively, were made into three different sizes to examine the effect of coal size. Due to low combustion temperature and low burning efficiency of the volatile matter in household stoves, PMs emitted from household coal combustion often have a high fraction of carbonaceous compositions. Both $PM_{2.5}$ EF and the fraction of its carbonaceous constituents increase with an increase in the coal volatile matter content till about V_{daf} = 35% and then stay roughly flat. Addition of extraneous minerals results in a significant reduction of PM emissions. Increasing the coal size from ~1 cm to ~10 cm leads to a reduction of PM EFs by ~80%. The coal volatile matter content determines the total amount of organic compounds to be devolatilized during coal pyrolysis stage. The coal size and ash content affect the escape of these organic compounds from burning coal chunks or briquettes. Together, they all affect the burning completeness of these organic compounds in household stoves and subsequently the total amount of organic precursors to form carbonaceous constituents in PMs.

1. Introduction

Household coal combustion for heating and cooking, especially in developing countries such as China and India, accounts for a large fraction of the global emissions of anthropogenic primary particulate matter (PM) and black carbon (BC) [1–4]. Owing to incomplete combustion and with no air pollution control device installed, residential coal combustion has significantly high emission factors (EFs) of PM, elemental carbon (EC), and organic carbon (OC) [5–13]. These high emissions from residential coal combustion are concerned due to the negative health impact associated with $PM_{2.5}$ (PM with aerodynamic diameter less than or equal to 2.5 μm) [14–18], as well as the negative impact on climate and visibility [19,20]. During high PM pollutant episodes in winter, coal combustion is possibly the largest anthropogenic source, especially for organic aerosol [21–23]. In response to haze pollution and visibility degradation in China, the government has updated policies on residential coal properties to reduce PM emissions from residential activities, e.g., lowering the threshold values for coal ash on dry basis (A_d, required to be less than 16% for

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Beijing-Tianjin-Hebei region) and coal volatile matter content on dry and ash-free basis (V_{daf}, required to be less than 10% and 20% for anthracite and bituminous briquettes, respectively, in Beijing) [24,25]. In 2014, Beijing government spent ~120 million USD as financial subsidy for coal replacement in residential heating application. However, limited work has been performed on the roles of residential coal properties in $PM_{2.5}$ emission from household combustion. For instance, no published results are available to support the current policy on coal ash content requirement for household application. Efforts to reduce $PM_{2.5}$ emissions from household activities will be hindered if coal properties are not properly addressed.

The coal volatile matter content was widely reported to influence PM EFs and their carbonaceous compositions [1,7,8,10,12,26,27]. However, previous results on the relationship between PM EFs and V_{daf} are inconclusive. Based on nine coal samples, EFs of PM, OC, and EC were reported to increase with an increase in V_{daf} from anthracites to bituminous coals with medium volatile matter content ($22\% < V_{daf} \leqslant 31\%$), and then decrease with further increase in V_{daf} for bituminous coals with high volatile matter content [7,12,28]. No detailed explanation and reliability analysis were provided for the above reported trend. Since the coal volatile matter and their products are main products to the incomplete combustion in household stove [10], PMs emitted from household coal combustion often have high fraction of carbonaceous compositions [5,12]. The reported decreasing trends of PM EFs and their carbonaceous compositions with the increase in V_{daf} for coals with high volatile matter content [7] are difficult to justify, when other factors such as stove and combustion condition were maintained the same. Experimental measurements on more coal samples with controlled properties, especially bituminous and lignite coals with high volatile matter content, will help to reveal this relationship.

Briquetting technology, simultaneously changing coal size and ash content, has been generally recognized to be a good approach to reduce PM emissions from household coal combustion [12,13,29]. The transformation of raw coal into briquettes includes steps such as pulverizing the coal, mixing it with binding material (e.g., bentonite clay and humic acid), and pressing the mixture into honeycomb or cake shapes [1,13,30,31]. Comparing to coal chunk (~3–10 cm in size), honeycomb-coal briquette (commonly larger size, ~10 cm, and higher ash content, e.g., ~30% with clay additions) was reported to reduce emissions of PM, OC, and EC by ~30–80% in various stoves [12,13,29]. The reduction benefitted from briquetting technology was briefly explained as that devolatilized matter are taken up by the clay binder before escaping from the solid matrix, and the clay serves as catalyst to crack down the coal tar into carbon and hydrogen [1,30]. This contradicts the current regulations in China that set an upper limit for residential coal ash. It is possible that current regulations are based on results from industrial coal fired boilers, which have much higher combustion temperatures than residential stoves. In these industrial boilers, an increase in coal ash (mineral matter) often results in increasing PM emissions due to the transformation of mineral matter into fly ashes during combustion process [32]. When developing regional and national PM emission inventories from industrial coal fire boilers, PM EFs are often assumed to positively correlate with coal ash content [32]. Different from industrial boilers, however, coal combustion in the household stove has a low combustion temperature and a low combustion efficiency [33]. PMs formed and emitted from the household coal combustion often have a low fraction of mineral compositions and a high fraction of carbonaceous species, though systematic investigations on the influence of coal ash content and coal size in household PM emissions are generally lacking.

Aiming to reveal the influences of coal size, volatile matter content, and additive in primary PM emissions, this study simulated household practices and characterized PM emissions by burning coal samples with controlled properties in a typical household stove. Effect of coal volatile matter content on $PM_{2.5}$ EFs and carbonaceous compositions were studied via burning fourteen coal chunk samples. Effect of various mineral additives on PM EFs was examined via burning briquette samples with different additives, while the effect of coal size was comparatively investigated by controlling chunk sizes. The influences of these three coal parameters on PM emissions were analyzed to reveal their roles in PM formation during the household coal combustion. Implications for reducing household PM emissions via coal property control were discussed.

2. Materials and methods

2.1. Coal samples and household stove

To examine the relationship between primary $PM_{2.5}$ EFs and V_{daf}, fourteen coal chunk samples were tested. These samples cover a wide range of geological maturities with V_{daf} ranging from 2.8% to 48.7% (see coal quality information in Table 1 in the Supporting Information). Contents of moisture, ash, fixed carbon, and volatile matter were obtained using the standard method of GBT 30732-201 and analyzed at China National Coal Quality Supervision Testing Center. These coal samples can be roughly classified into three anthracite samples with $V_{daf} < 10\%$, seven bituminous samples with $10\% < V_{daf} < 40\%$, and four lignite samples with $V_{daf} > 40\%$. These fourteen coal chunks have the size of ~3–10 cm in diameter, a typical size used in household stoves.

To investigate the influence of coal size on PM emissions, an anthracite chunk (V_{daf} = 9.26%, sample A3 in Table 1) and a bituminous chunk (V_{daf} = 37.69%, sample B10 in Table 1) were made into three size ranges, i.e., 0.8–2 cm, 4–7 cm, and 8–12 cm. Since the size and shape of coal chunks are not readily controlled, our investigation on the coal size effect is based on these three rough size ranges.

To investigate the influence of coal ash content on PM emissions, eight briquettes with various mineral matter additives were made into the same size and the same shape using the same manufacturing process and the same coal powder (sample B15 in Table 1). The coal powder was mixed with the same amount of binding agent (3%), water (10%), and different additives (at designated ratios). These mixtures were pulverized into finer powders with a quasi-uniform size (200–300 μm), and then pressed into coal cakes (~5 cm in diameter). SiO_2 powder (1–10 μm in diameter) was used as an additive with ratios of 5%, 10%, and 15% (mass basis). MgO powder (1–10 μm in diameter, 5%) and bentonite clay (5% and 10%) were also used as additives for three briquette samples. 2% bentonite clay was surface coated on the briquette with no other internally mixed additive as another briquette sample. One briquette sample with no additive was prepared for comparison. In practical briquette manufacturing, the bentonite clay is commonly used as the additive due to its viscous characteristics which facilitate the formation of coal cakes or honeycombs [30]. Bentonite clay was implied to potentially absorb devolatilized matter and to serves as a catalyst. In some applications, MgO and CaO are added in briquettes to reduce SO_2 emission [1,30]. SiO_2 powder was added in this study to test the effect of chemically inert minerals. All these eight types of briquettes were naturally dried and stored at the same place before the combustion experiment.

Combustion experiments for all the coal samples were performed in a steel stove (Sangpu, ZS60A), one of the most popular household stoves in rural Beijing. The coal weight was fixed at 3.0 kg for each combustion experiment. For the sake of consistency during the laboratory experiments and to avoid introducing