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Primary $PM_{2.5}$ and trace gas emissions from residential coal combustion: assessing semi-coke briquette for emission reduction in the Beijing-Tianjin-Hebei region, China

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ABSTRACT

In response to severe haze pollution, the Chinese State Council set $PM_{2.5}$ improvement targets for the Beijing-Tianjin-Hebei (BTH) region in 2013. To achieve the targets for the residential sector, semi-coke briquettes are being considered as a replacement for traditional raw coals with the help of financial subsidy, but information on the emission from them and the impacts on the air quality is limited. Laboratory experiments were conducted to determine emission factors (EFs) for a typical semi-coke briquette, its parent material (bituminous raw-coal-chunk) and three types of traditional coals (bituminous raw-coal-chunk, anthracite raw-coal-chunk and anthracite coal-briquette) extensively used in BTH. Compared with the parent material, significant lower EFs of primary $PM_{2.5}$, organic carbon (OC), element carbon (EC), the sum of 16 polycyclic aromatic hydrocarbon components (PAHs), SO_4^{2-}, NO_3^-, hazardous trace elements (HTEs) and NO_x were found in semi-coke briquette. A scenario for the BTH region in 2015 in which raw coals were replaced with the semi-coke briquette showed that amounts of pollutants emitted from residential coal combustion could decrease by 91.6% for primary $PM_{2.5}$, 94.0% for OC, 99.6% for EC, 99.9% for PAHs, 94.2% for NO_3^-, 45.6% for HTEs, 70.9% for NO_x and 22.3% for SO_2. However, SO_4^{2-} loadings evidently would increase if raw coals were replaced with either semi-coke briquette or anthracite coal-briquette. Geographic distributions of modeled reductions were developed to identify emission-reducing hot-spots and aid in the development of clean energy policies. Replacement of traditional raw coals with the semi-coke briquette apparently could lead to significant environmental improvements in BTH and other regions in China.

1. Introduction

China has been experiencing severe haze pollution, especially in winter when coal is used extensively for residential heating (Yan et al., 2017). Indeed, coal combustion is a major emission source of gases and fine particulate matter (PM, usually measured as $PM_{2.5}$, particles with aerodynamic diameters ≦ 2.5 μm). Emissions from coal combustion have led to severe indoor and outdoor air pollution, and they have become a serious national concern due to their adverse effects on health, visibility and the environment over regional and global scales (Huang et al., 2014; Li et al., 2017; Watson, 2002; Zheng et al., 2015).

The Beijing-Tianjin-Hebei (BTH) region is an important city agglomeration in China, where coal is still the primary fuel for residences in both urban and rural areas. In 2015, 20.5 Mt of raw coals were burned in BTH for residential uses, and that amounted to 20.3% of the total residential coal consumption in China (Fig. 1). Residential coal

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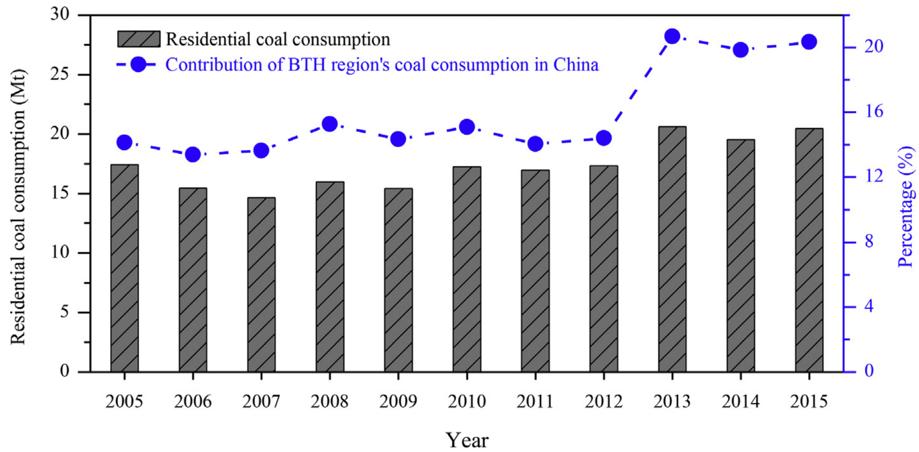

Fig. 1. Residential coal consumption in the Beijing-Tianjin-Hebei (BTH) region and the percentage of coal consumption in BTH out of the total household coal consumption in China. Source: China Energy Statistical Yearbook (2006–2016).

combustion is a major contributor to severe haze pollution in winter in this region. For instance, Zhang et al. (2017) reported that residential coal combustion contributed 46% of the monthly average $PM_{2.5}$ concentrations during BTH haze episodes in 2015 using WRF-CMAQ model. In response to the serious haze pollution events, the Chinese State Council released the National Air Pollution Prevention and Control Action Plan (2013–2017) in 2013 aiming to reduce $PM_{2.5}$ loadings. The policies were designed to cut down coal consumption and replace low rank coals (bituminous coals) with clean fuels as a way of reducing emissions from residential sources (Chinese State Council, 2013).

Semi-coke briquettes are industrial by-products made from bituminous raw-coal-chunks through a low-temperature carbonization process. These briquettes are being considered as a possible replacement of raw coals for residential usage (MEP, 2016), but knowledge on the emission from them is very limited (Li et al., 2016a, 2016b), and this complicates the evaluation process. Li et al. (2016a, 2016b) reported emission factors (EFs) of primary $PM_{2.5}$, particulate carbon, benzo [a] pyrene equivalent carcinogenic potency (BaP_{eq}), carbon monoxide (CO), carbon dioxide (CO_2) and sulfur dioxide (SO_2) emissions from household use of semi-coke briquettes. However, information on emissions of water-soluble ions, elements and nitrogen oxides (NO_x = NO + NO_2, a gas precursor of secondary $PM_{2.5}$) from semi-coke briquettes has not been obtained, and that information is needed to fully assess the feasibility of replacing raw coals with semi-coke briquettes.

In this study, we documented the EFs of primary $PM_{2.5}$ and selected chemical components, as well as trace gases including NO_x and SO_2 from residential coal combustion using custom-made combustion chamber. Seven coal samples were used in the tests, including one typical semi-coke briquette, its parent material and five traditional coals widely used in BTH. The major objective for the study was to estimate the emission reductions in BTH that might be achieved by replacing raw coals with semi-coke briquettes. To do this, the total emission reductions of primary $PM_{2.5}$ and trace gases were calculated, and the spatial distributions of the effects were modeled.

2. Experimental section

2.1. Fuel and stove

Selected properties by proximate and ultimate analysis of the seven coals tested in our study are presented in Table 1. The analyses of the materials were performed by Shaanxi Coal Geological Laboratory Co., Ltd., China using the national standards of the People's Republic of China (GB/T 211–2007, GB/T 212–2008 and GB/T213-2008). Semi-coke briquettes, which are the focus of this study, are made from the unprocessed

Table 1
Coal properties by proximate and ultimate analysis.

Type	Semi-coke	Bituminous			Anthracite	Anthracite	
	briquette	raw-coal-chunk			raw-coal-chunk	coal-briquette	
Coal ID	S-1	Bc-1[a]	Bc-2[b]	Bc-3[b]	Ac-1[b]	Ab-1[b]	Ab-2[b]
proximate analysis (wt%)							
moisture*	4.26	4.56	7.98	19.47	4.06	3.00	4.08
ash*	14.06	10.82	7.98	10.56	11.06	32.34	35.98
volatile matter*	4.32	31.26	33.2	24.56	7.22	4.99	5.04
fixed carbon*	77.36	55.81	50.84	45.41	77.66	59.67	54.90
calorific value (MJ/kg)**	24.01	24.87	25.82	20.02	28.56	20.37	18.48
ultimate analysis (wt%)							
C*	75.08	75.39	67.38	56.61	76.75	58.84	53.88
H*	0.76	4.07	3.74	2.26	2.28	0.84	0.80
O*	4.85	12.11	11.80	9.77	4.64	4.18	4.20
N*	0.58	0.92	0.94	0.67	0.86	0.48	0.67
S*	0.41	0.29	0.18	0.66	0.35	0.32	0.39

* based on air-dry basis.
** as received.
[a] The parent material used for producing semi-coke briquette (S-1).
[b] Traditional coals used in the Beijing-Tianjin-Hebei (BTH) region.

bituminous raw-coal-chunks by a low temperature coal carbonization process (500–800 °C) in a pyrolyzing furnace. Various substances in the coal vaporize or turn into liquid-phase tar and solid-phase semi-coke at specific temperatures. We collected one semi-coke briquette (S-1) and its parent material (bituminous raw-coal-chunk, Bc-1) from Yulin city, where is the biggest producing bases of semi-coke briquettes in China. Five coal samples were also purchased at local markets in BTH to obtain regionally representative coals used in household; these were two bituminous raw-coal-chunks (Bc-2 and Bc-3), one anthracite raw-coal-chunk (Ac-1) and two anthracite coal-briquettes (Ab-1 and Ab-2).

The test stove in this study was of a type widely used in Northern China for residential cooking and heating, and it was bought from a local market. The stove was 50 cm high, and its hearth had an outer diameter of 24 cm and an inner diameter of 12 cm. There was a 6 cm diameter air-control lip near the bottom, which was fully open during the combustion experiments to allow the maximum amount of air to enter the stove (Fig. S1).

2.2. Sampling and analysis

The experiments were conducted in a combustion chamber system