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8 AUTHORS, INCLUDING:


Dongyan Liu
Chinese Academy of Sciences


Gan Zhang
Chinese Academy of Sciences


Jun Li
Chinese Academy of Sciences

Guoying Sheng
Chinese Academy of Sciences

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Measurements of Black and Organic Carbon Emission Factors for Household Coal Combustion in China: Implication for Emission Reduction

YINGJUN CHEN,*,† GUORUI ZHI,‡ YANLI FENG,§ DONGYAN LIU,† GAN ZHANG,‖ JUN LI,‖ GUOYING SHENG,‖ AND JIAMO FU‖

Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai, Shandong Province 264003, China, Key Laboratory for Atmospheric Chemistry, Centre for Atmosphere Watch & Services of CMA, Chinese Academy of Meteorological Sciences, Beijing 100081, China, Institute of Environmental Pollution and Health, School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China, and State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China

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Household coal combustion is considered as the greatest emission source for black carbon (BC) and an important source for organic carbon (OC) in China. However, measurements on BC and OC emission factors (EF_{BC} and EF_{OC}) are still scarce, which result in large uncertainties in emission estimates. In this study, a detailed data set of EF_{BC} and EF_{OC} for household coal burning was presented on the basis of 38 coal/stove combination experiments. These experiments included 13 coals with a wide coverage of geological maturity which were tested in honeycomb-coal-briquette and raw-coal-chunk forms in three typical coal stoves. Averaged values of EF_{BC} are 0.004 and 0.007 g/kg for anthracite in briquette and chunk forms and 0.09 and 3.05 g/kg for bituminous coal, respectively; EF_{OC} are 0.06 and 0.10 g/kg for anthracite and 3.74 and 5.50 g/kg for bituminous coal in both forms, respectively. Coal maturity was found to be the most important influencing factor relative to coal's burning forms and the stove's burning efficiency, and when medium-volatile bituminous coals (MVB) are excluded from use, averaged EF_{BC} and EF_{OC} for bituminous coal decrease by 50% and 30%, respectively. According to these EFs, China's BC and OC emissions from the household sector in 2000 were 94 and 244 gigagrams (Gg), respectively. Compared with previous BC emission estimates for this sector (e.g., 465 Gg by Ohara et al., *Atmos. Chem. Phys.* 2007, *7*, 4419−4444), a dramatic decrease was observed and was mainly attributed to the update of EFs. As suggested by this study, if MVB is prohibited as household fuel together with further promotion of briquettes, BC and OC emissions in this sector will be reduced by 80% and 34%, respectively, and then carbonaceous emissions can be controlled to a large extent in China.

Introduction

Carbonaceous aerosols have generated wide concern in recent years due to their significant impacts on global and regional climate changes together with negative effects on the environment and human health (e.g., refs *1−3*). Carbonaceous aerosols are mainly derived from incomplete combustion of biomass and fossil fuels, and can be simply divided into black carbon (BC) and organic carbon (OC) fractions (*4*). Most concern about BC aerosol is due to its strong solar absorptivity, which contributes to global warming by increasing the top-of-atmosphere radiative forcing and decreasing the surface albedo of snow and ice, etc (*5−7*). Furthermore, BC aerosols reduce atmospheric visibility, damage the appearance of buildings, and do harm to human health by deeply penetrating into the lungs (*3*, *8*, *9*). OC has the optical property of mainly scattering solar radiation, but its ratio to BC affects the radiative property of BC (*10*). A large number of studies on OC have focused on their adverse effects on public health, because it contains hundreds of organic compounds and many species are toxic and carcinogenic (*11*).

BC emissions from China have caught great attention since Menon et al. (*12*) suggested the relationship between the increased BC aerosols and the precipitation trends in eastern China over the past several decades. Ramanathan et al. (*13*) recently identified one of hotspots of BC-induced atmospheric solar heating in this area. It was estimated that nearly 20% of global BC aerosol burden was originated from China (*4*), in which 35−45% might be derived from coal burning in the household sector (*14−17*). Household coal combustion also contributed a large proportion of OC emissions in China (*4*, *14−16*), and some organic compounds in coal smoke may be responsible for the notable lung-cancer mortality in Xuanwei, Yunnan Province, (*18*, *19*) and the high esophageal-cancer rate in Linxian, Henan Province (*20*). Unfortunately, there are still limited experimentally derived data about emission factors of BC and OC (EF_{BC} and EF_{OC}) in China, especially from household coal combustion, and this has resulted in high uncertainties in the estimates of carbonaceous emissions as well as difficulties in determining the best mitigation strategies (*4, 14−17, 21*).

EF_{BC} and EF_{OC} for household coal burning in China have been shown to be very difficult to measure experimentally (*22−27*). There are many factors which affect the formation of carbonaceous aerosols in coal smoke. These include mainly: (i) geological maturity of coal, from bituminous coal to anthracite, (ii) the burning form of coal such as raw-coal-chunk and honeycomb-coal-briquette, and (iii) burning efficiency of coal stoves. In our previous studies (*23−26*), various coals with a wide coverage of maturities were tested in form of chunk and briquette in representative coal stoves for the measurements of EF_{BC} and EF_{OC}. Significant impact has been shown about the coal's burning form and the stove's burning efficiency on the EFs (*26*), whereas the effect of coal maturity is emphasized in this study.

Therefore, the first purpose of this study is to present a detailed, experimentally derived data set of EF_{BC} and EF_{OC} for household coal combustion by considering various influencing factors mentioned above, together with annual

* Corresponding author. phone: +86-535-2109151; fax: +86-535-2109000; e-mail: yjchen@yic.ac.an.
† Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences.
‡ Chinese Academy of Meteorological Sciences.
§ Shanghai University.
‖ Guangzhou Institute of Geochemistry, Chinese Academy of Sciences.

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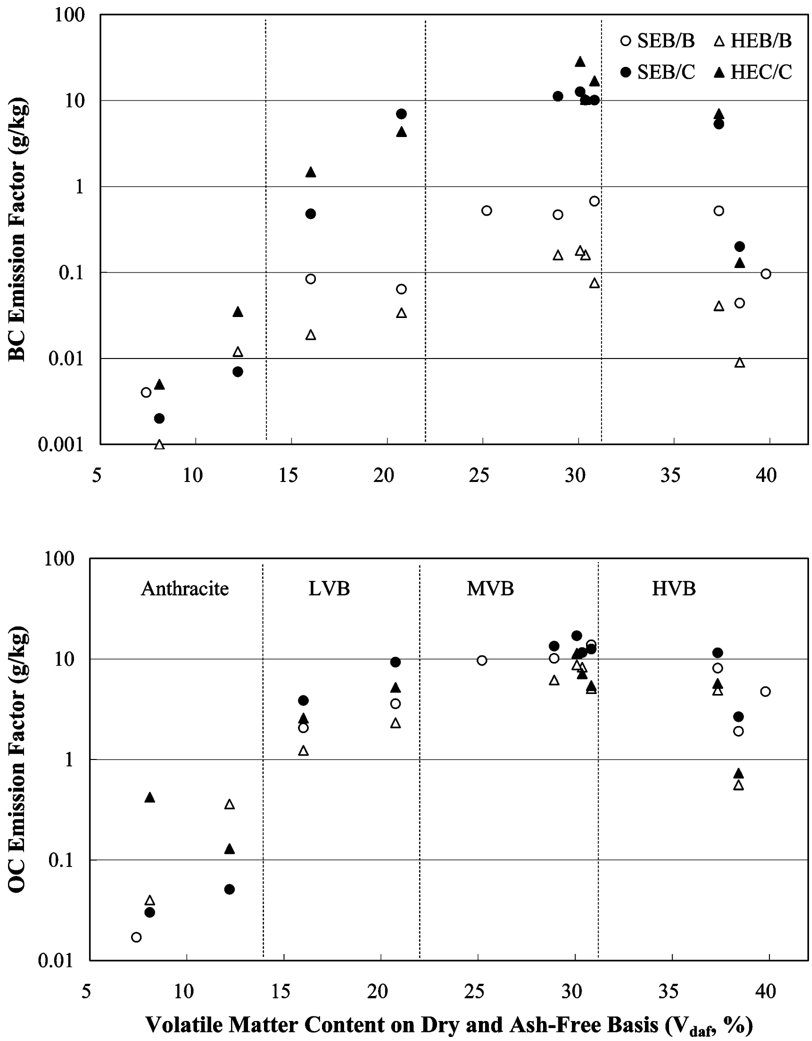

FIGURE 1. Variation trends of EF_{BC} and EF_{OC} with V_{daf} of coals under household burning conditions.

BC and OC emission estimates from this sector based on these EF data. The second purpose is to compare the importance of various factors influencing BC and OC emissions and discuss their significance for emission control in China.

Materials and Methods

Coal and Stove Combinations. Thirteen coal types were tested in our series of experiments (*23−26*), which covered a wide range of geological maturities and can be classified into three high V_{daf} (the volatile matter content on a dry and ash-free basis) bituminous coals (HVB), five medium V_{daf} bituminous coals (MVB), two low V_{daf} bituminous coals (LVB), and three anthracites. Three typical coal stoves with different burning efficiencies were selected for the experiments, including a simple and low-efficiency-briquette stove (SEB), a high-efficiency-briquette stove (HEB), and a high-efficiency-chunk stove (HEC). Coals were burned individually in chunk and honeycomb-briquette forms in the stoves. Detailed descriptions about the coals and stoves were presented in the Supporting Information. There were a total of 38 combinations of coal type/burning form/stove type tested in our experiments (Table S1 in the Supporting Information).

Sample Collection and Analysis. The sampling procedure and analytical protocol were described previously (*23−25, 28*) and also in the Supporting Information. Briefly, the sampling procedure started when coals (briquettes or chunks) were added into the stove and ignited by preburned charcoals and ended when the coals burned out without any disturbance. A sampling system gathered, diluted, and cooled down the coal smoke and ducted a fraction onto quartz fiber filter (QFF). Coal weights were recorded before and after combustion to calculate the actual burned mass, and the fractional ratio of collected on QFF to total emissions was monitored by two flow meters.

QFF samples were analyzed for elemental carbon (EC) and OC masses using a thermal-optical transmittance method (Sunset Laboratory, Inc., Forest Grove, OR) (*29*) with the temperature protocol described in the Supporting Information. Since a previous study had demonstrated that EC concentrations were comparable to BC measured by an optical method (Aethalometer AE90) for 11 coal/stove combinations (*25*), BC was considered as the same mass as EC in this study.

Results and Discussion

BC and OC Emission Factors. EF_{BC} and EF_{OC} for each coal/stove combination were calculated from BC and OC masses on the QFF sample, the fractional ratio of sampled to total emissions, and the actual burned coal weight (formula was provided in the Supporting Information). It should be noted that the ash content and unburned coal fraction were not taken into account in the EF calculations. Figure 1 contains all measurement results of EF_{BC} and EF_{OC} for 38 coal/stove combinations under household burning conditions (*23−26*). Similar bell-shaped trends are observed about these values with the V_{daf} of coals, and the highest values for each combination occur when V_{daf} is close to 30% or R_o is near

TABLE 1. Averaged EF_{BC} and EF_{OC} (g/kg) and Their Ratios for Household Coal Combustion in China

grouped coals	honeycomb-coal-briquette					raw-coal-chunk				
	EF_{BC}		EF_{OC}			EF_{BC}		EF_{OC}		
	GM[a]	SD[b]	GM	SD	BC/OC	GM	SD	GM	SD	BC/OC
anthracite	0.004	2.76	0.063	3.62	0.06	0.007	2.81	0.096	2.73	0.07
LVB coal	0.043	1.78	2.14	1.46	0.02	2.15	2.81	4.68	1.60	0.46
MVB coal	0.25	2.10	8.44	1.36	0.03	13.25	1.43	10.56	1.44	1.26
HVB coal	0.060	3.75	2.89	2.57	0.02	0.99	6.22	3.36	2.78	0.30
bituminous coal	0.087	1.38	3.74	1.33	0.02	3.05	1.82	5.50	1.34	0.55
bituminous coal without MVB	0.051	1.45	2.49	1.32	0.02	1.46	1.49	3.97	1.32	0.37

[a] Geometric mean. [b] Geometric standard deviation.

1.0%. Variations of several orders of magnitude in EF_{BC} and EF_{OC} values are observed among anthracites and various bituminous coals with different V_{daf}. For example, EF_{BC} ranges from 0.001 g/kg for AY coal in honeycomb-briquette form to 28.5 g/kg for CX coal in raw-chunk form, and EF_{OC} ranges from 0.017 g/kg for YX coal in briquette to 17.0 g/kg for CX coal in chunk (Figure 1 and Table S2 in the Supporting Information).

Geometric means of EF_{BC} and EF_{OC} were calculated for coal/stove combinations grouped by coal maturity and burning form, and great differences still exist among these groups (Table 1). For example, for bituminous coal, the mean EF_{BC} value decreases from MVB (0.25 g/kg) to HVB (0.06 g/kg) to LVB (0.04 g/kg) for briquettes, while the order was MVB (13.25 g/kg), LVB (2.15 g/kg), and HVB (0.99 g/kg) for chunks. The experiments by Zhang et al. (*27*) confirmed the high influence of the coal maturity on EF_{BC} and EF_{OC}. This means that accurately estimating the EF_{BC} and EF_{OC} for household coal burning is complicated, because coals are consumed in Chinese households with various maturities and burning forms.

In order to simplify the estimates of BC and OC emissions from household coal combustion, a set of EF_{BC} and EF_{OC} data is calculated for anthracite and bituminous coal in briquette and chunk forms separately (Table 1). For example, EF_{BC} values are 0.004 and 0.007 g/kg for anthracite in both forms, respectively, and 0.09 and 3.05 g/kg for bituminous coals (by averaging LVB, MVB, and HVB, but this process may contain uncertainty due to the actual fraction of each coal not being taken into account). Furthermore, MVB coals are relatively scarce and more often used for making coke than for household burning; if they are excluded from bituminous coals series, EF_{BC} and EF_{OC} values will decrease by about 50% and 30%, respectively.

In recent BC and OC emission inventories (e.g., refs *14* and *15*), the widely cited EF_{BC} and EF_{OC} data for household coal burning were derived from the compilation by Streets et al. (*4, 16, 17*). These data were calculated by multiplying the EF of particulate matter by the BC or OC fraction in particles from different studies, and the EF_{BC} values are 0.12 and 3.7 g/kg while the EF_{OC} values are 0.12 and 3.0 g/kg for anthracite or briquette and bituminous coal chunk, respectively. Compared with our results, these EF_{BC} data were close to but somewhat higher than the averages of bituminous coals; EF_{OC} were a little lower than the averages of bituminous coals excluding MVB (Table 1). The significant impact of coal maturity on the EFs was not considered in their calculations and, therefore, caused high uncertainties in their emission estimates (*30*).

Impact of Coal Maturity on EF_{BC} and EF_{OC}. It is necessary to clarify the impact of the various factors influencing EF_{BC} and EF_{OC} in order to control emissions in China. As mentioned above, there are three important factors relating to EF_{BC} and EF_{OC} for coal combustion in the household sector. Among them, the advantage of briquettes over chunks and improved stoves over simple stoves has been stressed previously (*26*). It was concluded that EF_{BC} and EF_{OC} and their ratios can be significantly lowered if all household coals are burnt in briquette form in HEB stoves. These results are corroborated by the present paper. For example, as showed in Table 1, averaged EF_{BC} for MVB in briquette is more than 50 times lower than in chunk form; the BC/OC ratio for bituminous coals reduces from 0.55 for chunks to 0.02 for briquettes, which may result in the coal smoke more optically scattering (*31*). However, these suggestions are difficult to carry out immediately in China, especially in remote rural areas due to poor traffic conditions and extra cost.

An alternative way for lowering EF_{BC} and EF_{OC} recommended in this study is to correctly choose the coal types as household fuel, because coal maturity was shown to have greater effects on EF_{BC} and EF_{OC} than the burning form and stove's efficiency under household burning conditions (Figure 1 and Table S2 in the Supporting Information). First, the four combinations of stove/burning form cannot change the bell-shaped trends of EF_{BC} and EF_{OC} with V_{daf}, although they have important effects on BC and OC emissions, especially for MVB. Second, coal maturity results in the difference of up to 3 to 4 orders of magnitude among EF_{BC} and EF_{OC} values, which is obviously greater than the impact of stove type/burning form combination. For example, the differences of the EF_{BC} and EF_{OC} values are about 6300 and 570 times between AY (V_{daf} is 8.1%) and CX coal (V_{daf} is 30.1%) chunks, respectively, whereas the differences are 70 and 2 times for CX coal between the two burning forms, respectively.

The strong dependence of EF_{BC} and EF_{OC} values on coal maturity is related to the poor conditions of household stoves, such as lower burning temperature, insufficient oxygen supply and mixture status, and shorter burning duration of volatile matter ejected when coal is heated. Coals with higher V_{daf} (i.e., HVB and MVB versus LVB and anthracite) are expected to produce more BC and OC aerosols under these conditions. However, unlike HVB coal whose volatile matter consists of a higher percentage of aliphatic homologues, MVB thermally releases a higher ratio of polycyclic aromatic hydrocarbons (PAHs) (*32*). Radke et al. (*33*) showed that MVB with R_o close to 0.9% yields the most abundant tar (mainly contains PAHs and their derivatives). The coal tar composes the majority of OC in the smoke (*23*) and is also the precursor of BC (*34*). Previous studies have elucidated that coal tar is eminently suitable for BC formation at the lower temperature in household stoves (ref *4* and references therein). Thus, it is fundamentally difficult to control the high emissions of BC and OC from MVB under household burning conditions.

BC and OC Emission Estimates. Using the EF data set from this study, combined with statistical data such as coal consumption, structure, and percentage of honeycomb briquette used, etc., we calculated annual BC and OC emissions for household coal burning in China. Table 2 presents the emission estimates for 2000 and a projection for 2020, with detailed discussion as follows.

TABLE 2. BC and OC Emission Estimates for Household Coal Combustion in China

		bituminous coal		anthracite		
		briquette	chunk	briquette	chunk	total
2000 yr	coal consumption (Mt)	25.30	37.95	6.33	9.49	79.07
	EF_{BC} (g/kg)	0.070	2.44	0.003	0.006	
	EF_{OC} (g/kg)	2.99	4.40	0.050	0.077	
	BC emissions (Gg)	1.77	92.61	0.02	0.06	94.45
	OC emissions (Gg)	75.65	167.0	0.32	0.73	243.7
2020 yr	coal consumption (Mt)	51.20	12.8	12.80	3.20	80.00
	EF_{BC} (g/kg)	0.046	1.31	0.004	0.006	
	EF_{OC} (g/kg)	2.24	3.57	0.057	0.086	
	BC emissions (Gg)	2.36	16.77	0.05	0.02	19.19
	OC emissions (Gg)	114.7	45.70	0.73	0.28	161.4

(1) Annual coal consumption in China has increased continually in recent years from 1.32 billion tons (Bt) in 2000 to 2.39 Bt in 2006; however, the household fraction varied in a narrow range of about 80 million tons (Mt) (*35*). Therefore, it is reasonable to predict the household coal consumption of 80 Mt will continue until 2020. In raw coal production, bituminous coals and anthracites account for roughly 80% and 20%, respectively (http://www.cct.org.cn), and this proportion is projected in the household sector.

(2) Average EF_{BC} and EF_{OC} from all bituminous coals were used in the calculation for 2000, while the values excluding MVB were adopted for 2020 (Table 1). Furthermore, 10% of averaged ash content in coals (see Table S1 in the Supporting Information) together with 10% of unburned coal fraction was considered in the EFs for 2000, whereas only ash content was taken into account for 2020.

(3) Honeycomb briquettes have been popularized through China during last two decades as a clean coal technology. However, there is no convincing statistical data about the true percentage of briquette usage. This study adopts the previous estimate of 40% for 2000 and 80% for 2020 (*23*), although some uncertainty may be caused here.

As shown in Table 2, annual BC emissions from household coal burning in China are 94 and 19 gigagrams (Gg) in 2000 and 2020, respectively. The 80% decrease is mainly attributable to the exclusion of MVB together with the increase of

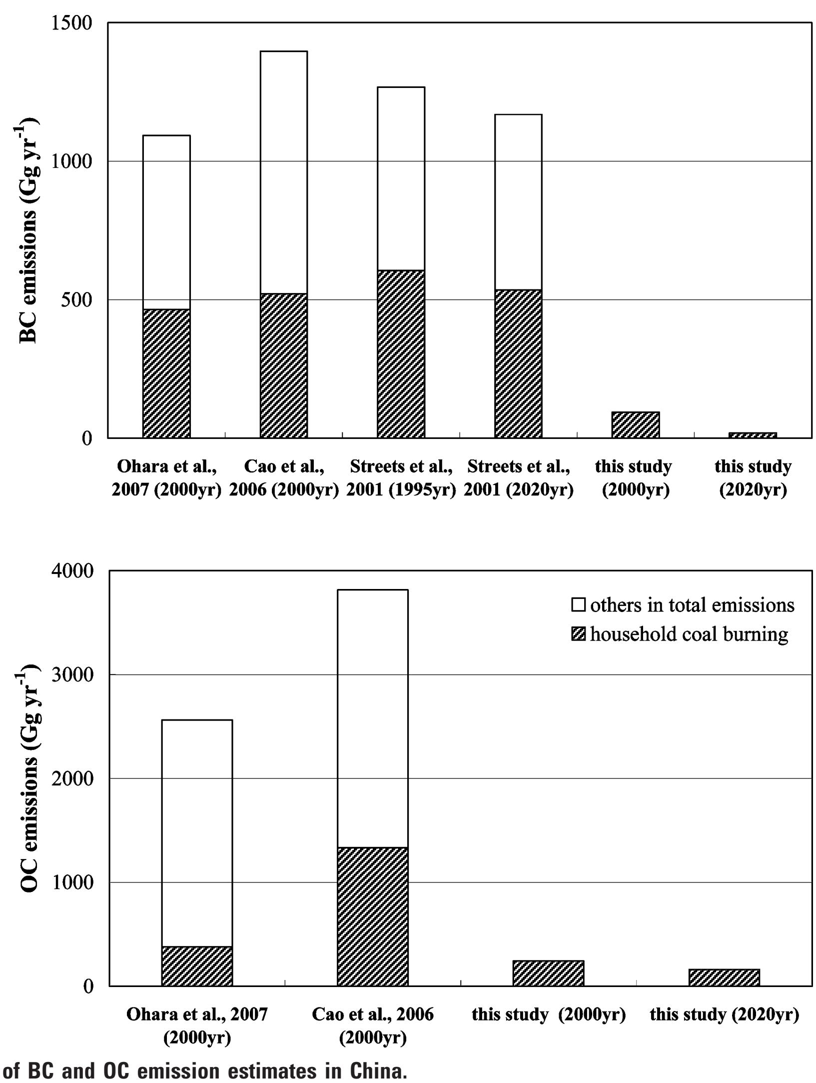

FIGURE 2. Comparison of BC and OC emission estimates in China.

briquette use. Bituminous coal chunk dominates the BC emissions although the contributing ratio decreases from 98% in 2000 to 87% in 2020 and, therefore, should be the first target for BC emission control.

By comparison, Ohara et al. (*14*) and Cao et al. (*15*) estimated the BC emissions of 465 and 521 Gg for this sector of China in 2000, respectively (Figure 2). The difference of 371 or 427 Gg between these data and our results is almost equivalent to the total BC emissions from North America (382 Gg) or Europe (466 Gg) in 1996 (*4*). Furthermore, according to Ohara et al. (*14*), BC emissions in 2000 were 1093 and 795 Gg from China and India, respectively, which were the biggest contributors in Asia. If 371 Gg is deducted from the BC emission inventory by Ohara et al. (*14*), China may be less than India for BC emissions, and this trend will be more obvious in 2020. On the other side, household coal combustion was estimated to be the dominant contributor (about 40%) of total BC emissions from China (1049 to 1396 Gg in 2000) (*14*−*16*), but its contribution is less than 10% according to our calculation. Therefore, on the basis of the EF_{BC} data set from experimental measurements, our studies may decrease some uncertainties in previous BC emission estimates from China.

The case for OC emissions is somewhat different from BC. The annual estimates of OC emissions in this sector are 244 and 161 Gg in 2000 and 2020, respectively. The OC emissions decrease by only 1/3 compared with the notable reduction for BC, and the OC to BC emission ratio increases by a factor of 3.3. Bituminous coal burning absolutely dominates the OC emissions in both years, although the contribution of coal chunks shifts from 69% to 28%. This is attributed to the minor difference of EF_{OC} between the two burning forms of bituminous coals.

There is significant discrepancy about the previous OC emission estimates in this sector and their contribution to total OC emissions in China (Figure 2). For example, Cao et al. (*15*) calculated 1333 Gg of OC emissions from household coal burning, which was the biggest contributor (35%) to the total emissions. However, the values decreased to 377 Gg or 15%, respectively, according to Ohara et al. (*14*), and then its contribution to total OC emissions was far lower than that of domestic biofuel consumption (82%). In spite of this, the estimate by Ohara et al. (*14*) is higher than our calculation by 55% and contains uncertainty due to EF_{OC} values adopted.

Implications for Emission Reduction. As discussed above, coal burning in the household sector is thought to contribute the most important fraction of BC and OC emissions in China. However, previous emission estimates contained significant uncertainties due to limited EF data. Our systematically designed experiments provide a set of EF_{BC} and EF_{OC} data grouped by maturity and burning form of coal. These data also provide plentiful information for BC and OC emission control strategies, including the correct choice of coal types (especially excluding MVB) for household use, the replacement of coal chunks by honeycomb briquettes, as well as the popularization of improved stoves. These measures are similar to the ones needed to decrease EF_{BC} and EF_{OC}, because coal cannot be completely replaced as household fuel in the coming decades.

Our previous study presumed that BC and OC emissions from household coal burning will be reduced by 98% and 61%, respectively, if all coal is burnt as briquettes in high-efficient stoves (*26*). However, according to the results in this study, the single measure of eliminating MVB from bituminous coals will decrease BC and OC emissions by 50% and 30%, respectively. Table 2 also shows that BC and OC emissions in 2020 will lower by 80% and 34% relative to 2000, respectively, when MVB is excluded and briquette usage rises from 40% to 80%. Furthermore, the exclusion of MVB from household fuel use will significantly benefit the indoor air quality and human health. A convincing example comes from the series of studies on the relationship of high lung-cancer mortality of women and children in Xuanwei, China, with the unvented burning of XW coal (MVB) (e.g., refs *18* and *19*).

In summary, coal maturity showed the most significant effects on BC and OC emissions in household coal burning, and MVB produced the highest values of EF_{BC} and EF_{OC} among various influencing factors. Therefore, for the benefit of both emission reduction and air quality improvement, a convenient way suggested by this study is to focus on carefully selecting bituminous coals and eliminating the use of MVB as household fuel. When these measures are carried out, together with further promotion of the use of honeycomb briquettes, total emissions of BC and OC in China will be significantly reduced.

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Supporting Information Available

A detailed description about the experiments and two tables separately containing basic information of various coal/stove combinations and EF_{BC} and EF_{OC} for an individual coal/stove combination are provided. This material is available free of charge via the Internet at http://pubs.acs.org.

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