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 to coal’s burning forms and the stove’s burning efficiency, (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 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...
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).
TABLE 1. Averaged EF_SC and EF_OC (g/kg) and their Ratios for Household Coal Combustion in China

<table>
<thead>
<tr>
<th>grouped coals</th>
<th>EF_SC</th>
<th>EF_OC</th>
<th>EF_SC</th>
<th>EF_OC</th>
<th>EF_SC</th>
<th>EF_OC</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>GM^a</td>
<td>SD^b</td>
<td>GM</td>
<td>SD</td>
<td>GM</td>
<td>SD</td>
</tr>
<tr>
<td>anthracite</td>
<td>0.004</td>
<td>2.76</td>
<td>0.063</td>
<td>3.62</td>
<td>0.06</td>
<td>2.93</td>
</tr>
<tr>
<td>LVB coal</td>
<td>0.045</td>
<td>1.78</td>
<td>2.14</td>
<td>1.74</td>
<td>0.06</td>
<td>2.81</td>
</tr>
<tr>
<td>MVB coal</td>
<td>0.25</td>
<td>2.10</td>
<td>8.44</td>
<td>1.36</td>
<td>0.02</td>
<td>1.43</td>
</tr>
<tr>
<td>HVB coal</td>
<td>0.060</td>
<td>3.75</td>
<td>2.89</td>
<td>2.57</td>
<td>0.02</td>
<td>3.00</td>
</tr>
<tr>
<td>bituminous coal</td>
<td>0.087</td>
<td>1.38</td>
<td>3.74</td>
<td>1.33</td>
<td>0.02</td>
<td>1.46</td>
</tr>
<tr>
<td>bituminous coal without MVB</td>
<td>0.051</td>
<td>1.45</td>
<td>2.49</td>
<td>1.32</td>
<td>0.02</td>
<td>1.49</td>
</tr>
</tbody>
</table>

a Geometric mean. b Geometric standard deviation.

1.0%. Variations of several orders of magnitude in EF_SC and EF_OC values are observed among anthracites and various bituminous coals with different Vdaf. For example, EF_SC 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 YY 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_SC 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_SC 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 Vdaf, 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 (Vdaf is 8.1%) and CX coal (Vdaf is 30.1%) chunks, 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 Vdaf (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 Rv 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.
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.

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.

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
EFBC data set from experimental measurements, our studies according to our calculation. Therefore, on the basis of the by 55% and contains uncertainty due to EFOC values adopted.

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 BC emissions 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), but its contribution is less than 10% according to our calculation. Therefore, on the basis of the EFBC 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 shifted from 69% to 28%. This is attributed to the minor difference of EFOC 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 EFOC 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 EFBC and EFOC 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 EFBC and EFOC, 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 MVF 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 MVF is excluded and briquette usage rises from 40% to 80%. Furthermore, the exclusion of MVF 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 MVF produced the highest values of EFBC and EFOC 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 MVF 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.

**Acknowledgments.** This work was supported by the National Natural Science Foundations of China (40605033, 40773047, and 40973071), Knowledge Innovative Programs of CAS (KZCX2-YW-Q07-04 and KZCX1-YW-06), Department of Science and Technology of Shandong Province (2007GZQZT018), and the Earmarked Fund of the State Key Laboratory of Organic Geochemistry (OGL-200804). We thank Dr. John Keesing and the anonymous reviewers for their help in revision.

**Supporting Information Available.** A detailed description about the experiments and two tables separately containing basic information of various coal/stove combinations and EFBC and EFOC for an individual coal/stove combination are provided. This material is available free of charge via the Internet at http://pubs.acs.org.

**Literature Cited.**


ES9021766