

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/7918039>

Emission Factors for Carbonaceous Particles and Polycyclic Aromatic Hydrocarbons from Residential Coal Combustion in China

ARTICLE *in* ENVIRONMENTAL SCIENCE AND TECHNOLOGY · APRIL 2005

Impact Factor: 5.33 · DOI: 10.1021/es0493650 · Source: PubMed

CITATIONS

172

READS

81

6 AUTHORS, INCLUDING:



Yingjun Chen

Tongji University

68 PUBLICATIONS 1,317 CITATIONS

SEE PROFILE



Guoying Sheng

Chinese Academy of Sciences

479 PUBLICATIONS 12,852 CITATIONS

SEE PROFILE



Xinhui Bi

Chinese Academy of Sciences

100 PUBLICATIONS 3,659 CITATIONS

SEE PROFILE



Yanli Feng

Shanghai University

23 PUBLICATIONS 893 CITATIONS

SEE PROFILE

Emission Factors for Carbonaceous Particles and Polycyclic Aromatic Hydrocarbons from Residential Coal Combustion in China

YINGJUN CHEN, GUOYING SHENG,
XINHUI BI, YANLI FENG,
BIXIAN MAI, AND JIAMO FU*

State Key Laboratory of Organic Geochemistry,
Key Laboratory of Guang Dong for Utilization and Protection
of Environmental Resources, Guangzhou Institute of
Geochemistry, Chinese Academy of Sciences,
Guangzhou 510640, People's Republic of China

Emission factors of carbonaceous particles, including black carbon (BC) and organic carbon (OC), and polycyclic aromatic hydrocarbons (PAHs) were determined for five coals, which ranged in maturity from sub-bituminous to anthracite. They were burned in the form of honeycomb briquettes in a residential coalstove, one of the most common fuel/stove combinations in China. Smoke samples were taken through dilution sampling equipment, with a high volume sampler that could simultaneously collect emissions in both particulate and gaseous phases, and a cascade impactor that could segregate particles into six fractions. Particulate BC and OC were analyzed by a thermal-optical method, and PAHs in emissions of both phases were analyzed by GC–MS. Burning of bituminous coals produced the highest emission factors of particulate matter (12.91 g/kg), BC (0.28 g/kg), OC (7.82 g/kg), and 20 PAHs (210.6 mg/kg) on the basis of burned dry ash-free (daf) coal, while the anthracite honeycomb-briquette was the cleanest household coal fuel. The size-segregated results show that more than 94% of the particles were submicron, and calculated mass median aerodynamic diameters (MMAD) of all particles were under 0.3 μm . Based on the coal consumption in the residential sector of China, 290.24 Gg (gigagrams) of particulate matter, 5.36 Gg of BC, 170.33 Gg of OC, and 4.72 Gg of 20 PAHs mass were emitted annually from household honeycomb-briquette burning during 2000. Anthracite coal should be selected preferentially and more advanced burning conditions should be applied in domestic combustion, from the viewpoint of both climate change and adverse health effects.

Introduction

There is considerable concern about carbonaceous aerosol emissions from coal combustion in China, due to its potential influence on climate change (1) and adverse effects on human health (2). About 1.2 billion tons of coal is consumed annually in China, equivalent to approximately 70% of the primary energy consumption, and more than 80% is burned directly (3). Therefore, roughly one-fourth of the global anthropogenic

carbonaceous emissions is believed to originate from China, of which about 70% is due to coal burning (4).

Carbonaceous particles are usually divided into two fractions, black carbon (BC) and organic carbon (OC). BC has a strong absorptivity of solar radiation (5) and therefore contributes to significant climate effects (6). Menon et al. (7) suggested that precipitation trends in China over the past decades, with increased rainfall in the south and drought in the north, might be related to increased BC aerosols. BC also contributed to the marked degradation of optical depths and visibility in northern China (8) and lowered the crop yields in China by reducing solar radiation that reaches the earth (9). In addition, BC can have adverse effects on human health (10), because fine BC particles can adsorb toxic pollutants and penetrate deeply into the lungs (11). OC has the optical property of mainly scattering solar radiation, and the ratio of OC to BC may affect the degree of radiative forcing of the atmosphere (12). Furthermore, OC often contains toxic organic compounds, such as polycyclic aromatic hydrocarbons (PAHs) and their alkylated homologues, which are noted carcinogens (13) and mutagens (14).

Some estimates had been made on the carbonaceous aerosol emissions from coal combustion in China, and these differed greatly among each other (4, 15–17). These differences derived primarily from the different emission factors assumed. Unfortunately, to our knowledge, few accurate measurements on carbonaceous emission factors from coal burning have been reported based on experiments. The emission factor of BC (EF_{BC}) was usually calculated indirectly by multiplying the emission factor of particulate matter (EF_{PM}) by the BC fraction of particles (F_{BC}), while few F_{BC} values from coal combustion were determined directly; therefore, the uncertainties of BC emission estimates were very high (15). The emission factor of OC (EF_{OC}) was estimated from the EF_{BC} and heterogeneous ratio of BC/OC, and different values were assumed (4, 18). In fact, EF_{BC} and EF_{OC} can vary greatly with coal/stove types, combustion conditions, and even burning stages (19).

The primary goal of this paper is to accurately quantify the emission factors of carbonaceous aerosol, including BC and OC, and PAHs from residential coal combustion in China. The residential sector is selected preferentially for two reasons. First, residential coal burning contributes the greatest part of the total carbonaceous emissions because of poor combustion conditions and lack of emission controls in China (15). Second, residential coal combustion results in the greatest indoor exposure to particles and PAHs, which may cause the most extensive health damage in China (20). In this work, five coals, covering a wide range of maturity, are chosen and burned in the form of honeycomb briquettes combined with domestic coalstove, which is advocated by the Chinese government as a clean-coal-combustion system and is expected to greatly reduce particulate emissions.

Experimental Section

Coalstove and Sampling System. The characteristics of the five coals selected are shown in Table 1. These five raw coals were all produced from state-owned coal mines and cover a wide range of maturity (vitrinite reflectance (R_o) from 0.52% to 2.50%), including a sub-bituminous (JP), three bituminous (XW, YM, and XA), and an anthracite (YX). These raw coals were powdered and intermixed with clay to produce honeycomb briquettes with a machine. The briquettes were 12-hole columns with a height of 6 cm and diameter of 9.5 cm.

The coalstove, the dilution sampling system, and the emission sampling procedure were described in detail

* Corresponding author phone: 86-2085290199; fax: 86-2085290192; e-mail: fujm@gig.ac.cn.

TABLE 1. Analytical Values of Five Raw Coals and Honeycomb Coal Briquettes^a

	JP	XW	YM	XA	YX
Proximate Analysis (wt (%), on air-dry (ad) basis)					
moisture (M_{ad})	3.33 (1.78) ^a	1.16 (0.82)	0.85 (0.28)	0.90 (0.37)	1.22 (1.44)
ash (A_{ad})	7.83 (38.07)	37.66 (50.53)	20.79 (45.84)	13.78 (39.62)	8.82 (52.91)
volatile (V_{ad})	35.55	27.82	23.30	21.47	6.68
fixed carbon (FC_{ad})	53.29	33.36	55.06	63.85	83.28
Elemental Analysis (wt (%), on dry ash-free (daf) basis)					
C_{daf}	74.59	78.64	82.65	85.01	90.58
H_{daf}	4.73	5.12	4.65	4.21	3.54
N_{daf}	4.19	5.15	5.36	4.81	0.58
O_{daf}	16.22	10.24	5.72	4.22	1.92
S_{daf}	0.28	0.86	1.61	1.76	0.67
H/C (daf)	0.063	0.065	0.056	0.050	0.039
O/C (daf)	0.217	0.130	0.069	0.050	0.210
vitrinite reflectance (R_o (%))	0.52	1.12	1.27	1.70	2.50

^a Analytical values for honeycomb coal briquettes are listed in parentheses after the corresponding raw coals, only for moisture and ash contents on an air-dry basis.

elsewhere (21) and are briefly summarized here. The coalstove is the most common household stove for burning honeycomb coal briquettes in China. It has a metallic outer cover and thermal-insulated ceramic liner and can hold three briquettes at one time. There is a 6 cm-diameter hole for air-control in the outer cover near the bottom. The sampling system was composed of a hood, three same-sized pipes, and a long curved pipe, with a total volume of 270 L. All parts were made of stainless steel, and Teflon was used for all gaskets to avoid organic contamination. The samplers used included a filter/PUF sampler, which could simultaneously collect particulate and gaseous organic matter using a glass fiber filter (GFF, 20 cm × 25 cm) and polyurethane foam (PUF, 6.5 cm diameter × 8 cm height) at the flow rate of 300 L/min, and a size-segregation sampler (HiVol cascade impactor, Thermo-Andersen Instrument Inc.), which could segregate particles into six fractions with cutoff diameters of >7.2, 3.0–7.2, 1.5–3.0, 0.95–1.5, 0.49–0.95, and <0.49 μm at a flow rate of 1.13 m³/min. The laboratory room designed especially for emissions testing had a filter system installed to provide only particle-free air.

Sampling Procedure. The honeycomb coal briquettes were ignited from the bottom using preweighed charcoal. To reduce the interference from the igniting fire, the charcoal was started with some paper and left to burn until the temperature in the stove was high and smoking stopped, and then the coal briquettes were placed into the stove. Sampling periods lasted for 1.5–4 h, which started when the coal briquettes were ignited and ended when the combustion finished completely. The weights of honeycomb briquettes before and after combustion were recorded to obtain the actual weight of coal burned, which varied from 50 to 350 g, and about 10% of ash-free coal remained unburnt. The air-control hole was fully opened to let the most air in during combustion.

All parts of the sampling system in contact with the flue gas were cleaned thoroughly before sampling, and the system was checked for leakage after assembly. During sampling, the hood was hung above the stove without touching to collect the emissions but not disturb the burning. The curved pipe was immersed into cool water, which further dropped the flue gas temperature to ambient (23–31 °C). The actual emission temperature in the flue was recorded every 10 min. Because the greatest emission rate of flue gas from the stove was measured to be 30 L/min, the dilution ratios of additional clean air to the emissions were at least 9:1 and 37:1, respectively, for the filter/PUF sampler and the cascade impactor, while the residence times of flue gas were about

54 and 14 s before being collected, respectively. No loss of emissions and minor deposits in the pipes were observed.

Analytical Methods. The analytical procedure for PAHs was given elsewhere (22), but a brief summary follows. Filter samples were extracted ultrasonically three times with 150 mL of dichloromethane (DCM), and each extraction lasted 30 min. PUF samples were extracted for 72 h with DCM in a Soxhlet apparatus. Surrogate deuterated PAHs were added prior to extraction. The extracts were concentrated on a rotary evaporator (BÜCHI Rotavapor R-114) and fractionated using silica–alumina column chromatography. The PAH fractions were concentrated using rotary evaporation followed by a gentle stream of nitrogen, and internal standard (hexamethylbenzene) was added for quantification of individual PAHs by GC–MS (HP-5890GC coupled with 5972MSD).

Portions of filter samples (1.5 cm²) were cut for analyzing organic and elemental carbon contents (OC/EC) by a thermal-optical carbon analyzer (Sunset Laboratory Inc., Forest Grove, OR) with a modified NIOSH (National Institute of Occupational Safety and Health) thermal-optical transmission (TOT) protocol (23). The punch aliquot of filter was heated stepwise at temperatures of 310, 475, 575, and 650 °C in a pure helium atmosphere for detecting the OC fraction, and at temperatures of 550, 625, 700, 775, and 850 °C in an oxidizing atmosphere of 2% oxygen (by volume) in a balance of helium for detecting the EC fraction. The carbon evolved at each temperature is oxidized to carbon dioxide (CO₂) and then reduced to methane (CH₄) for quantification with a flame ionization detector (FID). Some of OC pyrolyzes to EC as the temperature increases in inert helium and is subtracted from the elemental carbon area according to the initial laser absorbance. At the end of every analysis, a fixed volume loop of methane is injected automatically as an internal standard to calculate the carbon results.

Quality Control. Field blanks and charcoal emission samples were collected to determine background contamination. As compared to coal emission samples, filter samples for field blanks and charcoal emissions contained very low particles and PAHs, while PUF samples of charcoal emissions contained about 15% of the PAH concentrations of the coal briquette emission samples. Therefore, the individual PAH concentrations in charcoal PUF samples were subtracted from coal emission samples. Surrogate standards were added to all of the samples (including field blanks and charcoal emissions) to monitor procedural performance and matrix effects. The recoveries for deuterated PAHs ranged from 84% to 105%, and PAH concentrations were corrected by the recovery efficiency of the deuterated PAHs.

TABLE 2. Emission Factors (g/kg) of Particulate Matter, Black Carbon, and Organic Carbon from Honeycomb Briquette Combustion of Five Coals Based on Burned Dry Ash-Free Coal Weight

	sub-bituminous		bituminous			anthracite
	JP	XW	YM	XA	geomean	YX
EF _{PM}	8.001	19.550	14.134	7.788	12.911	1.329
EF _{BC}	0.096	0.675	0.523	0.064	0.283	0.004
EF _{OC}	4.729	13.818	9.656	3.580	7.817	0.017
OC/BC	49.312	20.483	18.474	56.085	27.686	4.807
OC/PM (%)	59.11	70.68	68.32	45.97	60.55	1.31
BC/PM (%)	1.20	3.45	3.70	0.82	2.19	0.27

The reproducibility of the collecting procedure was checked in three separate experiments for the filter/PUF sampler and in two experiments for the cascade impactor. The two sets of six-stage samples were combined for analysis, while the three sets of GFF/PUF samples were analyzed separately. All of the analytical results of PAHs in GFFs and PUFs are reported as the arithmetical mean values, because the relative standard deviations (RSD) were less than 20%. Three to five portions for each GFF sample were analyzed in the OC/EC analyzer to eliminate the nonuniformity of filter samples. The RSDs for OC and EC among three separate sets of filters were smaller than 15%. Therefore, the arithmetical mean values were calculated to represent the OC and EC results of various coal-burning emission samples.

Results and Discussion

Particulate Emissions. The emission factors of particulate matter (EF_{PM}) from honeycomb briquette combustion of five coals (based on burned dry ash-free coal (daf)) are presented in Table 2. There are pronounced differences across all of the coals. Bituminous coals yield the highest EF_{PM}, and the average value is 12.9 g/kg, while sub-bituminous and anthracite produce 8 and 1.3 g/kg of particles, respectively. Among the three bituminous coals, EF_{PM} of XW is the highest (19.6 g/kg).

It is accepted that coals with high volatile content have higher particulate emissions than coals with less volatiles. EF_{PM} for bituminous coal was found to be 20 times higher than that for anthracite burned in same stove (24). However, coal combustion involves a series of complicated chemical reactions, including pyrolysis, bond cleavage, and tar formation and further thermal oxidation, etc. (25, 26). Under residential burning conditions, coal tar may be released into the flue and form significant particulate matter (19). This is confirmed by fluorescence micrographs of filter samples in the present work, where many micro-drops of coal tar were observed on the filters, especially those from high volatiles bituminous coals. Coal tar yield has been found to be related to volatile content and coal rank. High volatile bituminous forms the most tar, followed by sub-bituminous and anthracite coal (27). On the other hand, Radke et al. (28) showed that bituminous coal yields the most tar when its R_o (vitrinite reflectance, a measure of the geological maturity of the organic matter) is around 0.9%. Therefore, as anticipated, among three bituminous coals in the present study, XW (R_o is 1.12%, the closest to 0.9%) yields the highest EF_{PM}, followed by YM and XA. The sub-bituminous coal (JP) only produces a EF_{PM} similar to that of XA, although its volatile content is the highest, and the anthracite (YX) yields the lowest value.

There are some EF_{PM} values for residential coal combustion published in other studies (19, 24, 29–33), which are generally similar to the present results. For example, Bond et al. (19) reported a value of 12 ± 17 g/kg for bituminous coal although it was burned as raw coal, very close to the present result (12.9 g/kg). Streets et al. (15) recently calculated the EF_{PM} values for residential coal burning from previous results and reported 8 g/kg for bituminous and 0.5 g/kg for

briquette or anthracite coal. However, honeycomb coal briquettes are thought to have reduced particulate emissions by 5–9 times as compared to raw coal (34). Zhang et al. (29) reported different EF_{PM} values of 8.73 and 0.27 g/kg for unprocessed coal and honeycomb briquette, respectively, using the same coal and stove, but the coal type and stove they used were different from that in the present work. The different values of EF_{PM} reported may be attributed to the different coals selected and the combustion conditions including stove types and burning temperatures (35), and the differences of EF_{PM} for various coal types and burning conditions may be obviously greater than that from briquetting. That is to say, the expected reduction of EF_{PM} caused by briquetting of coal may be offset by the differences due to coal maturities and combustion conditions. Therefore, the EF_{PM} data in this study were comparable with others. Sampling methods may also affect the measurements. In general, the emission rate during the first few minutes of coal ignition is up to 50 times higher than that during the remainder of the burning period (19, 24). As an example, a very low value of EF_{PM} (7 mg/kg) for coal briquette combustion was measured by Oanh et al. (30), who excluded the fire induction period from sampling to reduce contamination by supplementary fuels for ignition.

It should be noted that choked burning conditions (the inlet was partially blocked), which usually occur in domestic coal-use, are not included in the present experiments. However, high emissions can result from choked combustion. Bond et al. (19) obtained different values of 12 ± 17 and 9 ± 5 g/kg for sampling with and without inclusion of choked combustion, respectively. That is to say, the emission factors of particulate matter from the typical practice of residential coal combustion should be somewhat higher than these reported values.

Particle Size Distribution. The weights of particles collected on the six stage filters were recorded for calculating the mass percentage of each stage to total weight and the mass median aerodynamic diameter (MMAD, particle diameter where one-half of the particle mass is smaller and the other half is larger (36)) (Table 3). Submicron particles (sum of the fifth and sixth stages, aerodynamic diameter is $<0.95 \mu\text{m}$) dominate the particulate mass emitted from honeycomb coal briquettes combustion, and the calculated MMAD values are about $0.29 \mu\text{m}$. Factually, these freshly emitted particles should be finer if the cutoff size of the last stage (backup filter) of the size-segregated sampler is not limited to $0.49 \mu\text{m}$. Bond et al. (19) found that particles emitted from coal briquette and bituminous coal are largely in the nuclei mode (20–100 nm). For comparison, the results of field- and laboratory-scale studies on emissions from pulverized coal combustors indicated that the particle size distributions are typically bimodal, and the fine particle mode is around $0.1 \mu\text{m}$, which appears to result from vaporization-condensation processes (37, 38). This means that the fine particles from a domestic coalstove can penetrate deeper into the alveolar regions of the lung and pose greater threat to human health (39).

TABLE 3. Mass Percentage (%) for Particulate Fractions of Six Size Cuts to Total Emission Mass and Mass Median Aerodynamic Diameters (MMAD) in Honeycomb Briquette Combustion Emissions from Five Coals

coal	>7.2 μm	3.0–7.2	1.5–3.0	0.95–1.5	0.49–0.95	<0.49 μm	MMAD (μm)
JP	0.87	1.32	1.25	2.08	6.16	88.32	0.28
XW	1.03	1.86	1.86	2.77	7.83	84.65	0.29
YM	0.98	0.92	1.26	2.80	10.70	83.33	0.29
XA	1.03	1.14	1.03	1.99	12.73	82.08	0.30
YX	0.80	1.05	1.05	2.24	6.54	88.31	0.28

It has been found that the residence time of particles emitted into the atmosphere before being sampled and the dilution ratio of air to the emissions can significantly affect the total particle number concentration and size distribution, and longer residence time will shift the particles to larger sizes while a greater dilution ratio will increase the concentration of ultrafine particles (40, 41). As mentioned above, the residence time and dilution ratio in the present experiments are about 14 s and 37:1, respectively, which are much too small as compared to the actual situation in domestic living. On other words, there are uncertainties about the particle size distribution in field combustion. However, a conclusion can be drawn at least that adverse health effects will occur when exposed in fresh emissions from residential coal combustion.

BC and OC Emission Factors. Black carbon (BC) and elemental carbon (EC) are terms describing the light-absorbing fraction of carbonaceous aerosol. They are often used interchangeably, although there are obvious differences between them. BC is measured using an optical absorption method, while EC is usually determined by a thermal-optical technique. Turpin et al. (42) compared photoacoustic spectroscopy with thermal-optical transmittance (TOT) when measuring atmospheric EC (they did not discriminate these terminologies) and found there was no significant difference between the two data sets at the 95% confidence level. Because the term of BC was used more frequently than EC when the carbonaceous particulate matter emitted from coal combustion was discussed, BC was selected in this paper and the masses of BC and OC on filter samples were thought to be equal to that of EC and OC analyzed by the NIOSH TOT protocol (23).

The emission factors of BC (EF_{BC}) and OC (EF_{OC}) for five coals (daf) in this work are given in Table 2. The variation tendencies of EF_{BC} and EF_{OC} among various coals are similar to that of EF_{PM} ; that is, bituminous coals have the highest values of EF_{BC} (0.28 g/kg) and EF_{OC} (7.82 g/kg), while anthracite yields the lowest emission factors of BC (4 mg/kg) and OC (17 mg/kg).

A good correlation is found between EF_{OC} and EF_{PM} ($R^2 = 0.99$) of these coals, which is due to the fact that PM emitted from bituminous and sub-bituminous coals consists mainly of coal tar, as mentioned above. The ratio of OC/PM (F_{OC}) is about 60%, and the maximum is 70% for XW (Table 2), and organic matter ($1.3 \times \text{OC}$) (18) contributes up to nearly 80% of PM mass. Similar data have been reported when the indoor levels of PM from smoky coal (XW) burned in open-pits were collected (43, 44). Because coal tar is thought to be the precursor to BC (45), good correlations of EF_{BC} versus EF_{OC} ($R^2 = 0.94$) and EF_{BC} versus EF_{PM} ($R^2 = 0.90$) are expected.

Oros and Simoneit (46) had reported EF_{BC} values of 1.4 g/kg for bituminous and 0.5 g/kg for sub-bituminous coal from laboratory-scale combustion, several times higher than the present results, while the EF_{OC} values were very close to our measurements. The different coal types and combustion conditions may contribute to the difference of EF_{BC} , and the reason may be similar to that for different EF_{PM} . As an example, the difference of EF_{BC} between YM and XA in this work is greater than 10 times (Table 2). When the flame is

not hot enough to burn volatilized organic matter, it is also too cool to produce BC (47).

However, other EF_{BC} and EF_{OC} values assumed for emission inventories are much higher and differ greatly with each other (4, 15, 16, 18, 48, 49), and this can be due to the different F_{BC} (the BC fraction of particles) values and BC/OC ratios they used. For example, although the EF_{BC} values calculated by Streets et al. (15) are much lower than other assumptions, the F_{BC} used (45% for bituminous and 24% for briquette) are higher as compared to the present results (Table 2). Lioussse et al. (18) presented the first estimate about OC emissions, and assumed EF_{OC} (33.3 g/kg) from EF_{BC} of 10 g/kg and a BC/OC ratio of 0.3, while Cooke et al. (4) chose a value of 1:2 for the BC:OC ratio and obtained a value of about 10 g/kg for EF_{OC} .

It should be noted that different analytical protocols for OC and EC are expected to give different results despite the similar thermal-optical technology. For example, the maximum temperature selected for OC analysis in the oxygen-free atmosphere usually varies from 550 to 900 °C and affects the results, but the reason remains unclear. Chow et al. (50) compared the IMPROVE (Interagency Monitoring of Protected Visual Environments) with the NIOSH protocols (23), and concluded that NIOSH EC was typically less than half of IMPROVE EC, and primarily attributed this difference to the different allocation of carbon evolving at the NIOSH 850 °C temperature in a helium atmosphere (51). However, Schauer et al. (52) suggested some organic compounds did not pyrolyze or volatilize when the maximum temperature in pure helium was too low, and therefore the EC value was overestimated. In the present experiments, the maximum temperature for detecting OC was 650 °C, which was suggested by Sunset Laboratory for analyzing the GFF samples.

PAH Emission Factors. The emission factors of 20 PAHs, including 14 parent PAHs and 6 alkylated homologues, based on burned coal-weight (daf) are presented in Table 4. It should be noted that the PAH emissions here are the sum of both particulate and gaseous phases, and the percentages of particulate phase to the sum are also listed in Table 4.

The combustion of bituminous and sub-bituminous coals produced 210.6 and 121.2 mg/kg of total 20 PAHs, while anthracite (YX) yielded an amount 3 orders of magnitude lower. The same tendencies were observed for some carcinogenic parameters other than sole BaP for the whole PAH fraction in emissions from various coal combustions (Table 4). These parameters include $\Sigma_{\text{gen-PAHs}}$ (sum of six carcinogenic PAHs, such as BaA, Chry, BbKF, BaP, DbahA, and IncdP, which are classified as Group B2 (13)), BaPE (BaP-equivalent carcinogenic power, introduced by Yassaa et al. (53, 54)), and $\Sigma\text{BaP}_{\text{eq}}$ (sum of PAHs multiplied by their toxic equivalency factors introduced by Nisbet and LaGoy (55)).

The PAH emission from various coals is found to have a relationship with their volatile content, and the complete combustion of coals with a high volatile content is more difficult to achieve (30). Anthracite coal contains very little volatile matter and can therefore burn more completely.

PAHs released from coal combustion are believed to consist of two categories according to their formation

TABLE 4. Emission Factors of PAHs from Honeycomb Briquette Combustion of Five Coals Based on Burned Dry Ash-Free Coal^a

PAHs ^b	sub-bituminous		bituminous						anthracite		
	JP (mg/kg)		XW (mg/kg)		YM (mg/kg)		XA (mg/kg)		geomean ^c	YX (μg/kg)	
	total	P% ^d	total	P% ^d	total	P% ^d	total	P% ^d	total	total	P% ^d
Flu	4.160	6.22	11.064	3.22	5.805	15.96	1.165	1.61	4.214	9.136	0.05
Phe	25.817	31.31	45.307	46.22	84.855	34.09	38.204	9.51	52.762	91.318	0.08
Ant	8.620	30.19	8.791	52.30	8.093	46.99	1.728	14.45	4.972	2.260	0.10
Fluo	6.302	92.18	6.436	98.90	8.603	94.07	3.495	81.72	5.784	5.215	1.47
Pyr	2.237	93.66	2.973	97.08	3.506	94.14	1.208	85.16	2.327	4.908	1.70
BaA	2.947	99.81	4.887	99.85	6.106	99.83	2.697	99.70	4.318	0.241	33.64
Chry	4.998	99.68	10.378	99.67	15.879	99.66	10.055	99.37	11.833	2.376	32.57
BbKF	5.251	100	3.862	100	6.942	100	5.279	100	5.211	2.669	85.17
BeP	2.511	100	4.880	100	7.649	100	4.260	100	5.418	2.065	92.20
BaP	1.613	100	2.155	100	2.458	100	1.048	100	1.771	0.190	100
IncDP	1.604	100	0.942	100	1.461	100	0.899	100	1.073	0.922	100
DbahA	0.833	100	1.595	100	3.161	100	1.994	100	2.158	0.657	100
BghiP	1.778	100	2.692	100	4.865	100	2.436	100	3.171	1.219	100
Cor	0.338	100	0.440	100	0.897	100	0.738	100	0.663	1.244	100
Mflu	10.537	14.33	33.772	14.61	27.827	20.94	4.508	4.47	16.180	1.914	0
C2-Flu	8.546	46.84	28.090	58.79	26.452	56.04	3.418	31.52	13.643	1.245	0
Mphe	16.476	69.23	54.456	82.69	65.343	69.78	14.858	40.10	37.532	3.386	0.46
Dmphe	9.988	88.32	41.339	96.24	34.758	89.09	5.376	75.84	19.768	1.629	0.75
Mpyr	1.901	98.99	4.583	99.28	4.043	98.64	1.135	97.52	2.760	0.275	8.91
Mchry	4.728	100	18.598	99.96	22.181	99.96	8.247	99.91	15.040	0.137	100
ΣParent-PAHs	69.009		106.40 3		160.28 1		75.206		105.676	124.42 0	
ΣAlkyl-PAHs	52.175		180.83 7		180.60 4		37.541		104.923	8.585	
ΣPAHs	121.184		287.24 0		340.88 4		112.74 7		210.599	133.00 5	
BaPE	2.785		3.751		5.324		2.847		3.775	0.859	
Σgen-PAHs	17.246		23.819		36.008		21.971		26.365	7.054	
ΣBaP _{eq}	3.619		5.004		7.461		4.115		5.254	1.399	

^a Unit of emission factors for sub-bituminous and bituminous honeycomb coal briquettes is mg/kg, while unit for anthracite coal is μg/kg.

^b Abbreviations of PAHs: Flu, fluorene; Phe, phenanthrene; Ant, anthracene; Fluo, fluoranthene; Pyr, pyrene; BaA, benz[*a*]anthracene; Chry, chrysene; BbKF, benzo[*b*+*k*]fluoranthene; BeP, benzo[*e*]pyrene; BaP, benzo[*a*]pyrene; IncDP, indeno[1,2,3-*c,d*]pyrene; DbahA, dibenz[*ah*]anthracene; BghiP, benzo[*ghi*]perylene; Cor, coronene; Mflu, methylfluorenes; C2-Flu, C2-fluorenes; Mphe, methylphenanthrenes; Dmphe, dimethylphenanthrenes; Mpyr, methylpyrenes; Mchry, methylchrysenes; ΣParent-PAHs, sum of 14 parent PAHs above; ΣAlkylated-PAHs, sum of six alkylated PAHs above; ΣPAHs, sum of all 20 PAHs listed; BaPE, benzo[*a*]pyrene-equivalent carcinogenic power (BaPE = BaA × 0.06 + BbKF × 0.07 + BaP + DbahA × 0.6 + IncDP × 0.08); Σgen-PAHs, sum of six carcinogenic PAHs (BaA, Chry, BbKF, BaP, DbahA, and IncDP); ΣBaP_{eq}, sum of individual PAHs multiplied by their toxic equivalency factors. ^c Geometric average of the results of three bituminous coals. ^d PAHs in particulate phase in percentage of total PAHs detected in both particulate and vapor phases.

TABLE 5. Emission Estimates for China of PM, BC, OC, and PAHs from Residential Combustion of Five Honeycomb Coal Briquettes during 2000

	percentage (%) ^a	consumption (Tg) ^b	emission factors ^c (g/kg)				emissions (Gg) ^d			
			EF _{PM}	EF _{BC}	EF _{OC}	EF _{PAHs}	PM	BC	OC	PAHs
anthracite	17.57	5.557	1.329	0.004	0.017	1.33 × 10 ⁻⁴	7.385	0.022	0.094	7.39 × 10 ⁻⁴
bituminous and sub-bituminous	78.07	24.693	11.455	0.216	6.894	1.913	282.85 8	5.334	170.23 4	4.723
total		31.629 ^e					290.244	5.356	170.328	4.723

^a Percentage of anthracite and bituminous coal in whole yield of raw coal in 2000 in China (refer to <http://www.cct.org.cn/cct/>). ^b Teragram or Mt. ^c Emission factors for bituminous are the geomean value of a sub-bituminous and three bituminous coals. ^d Gigagram or kilotonnes. ^e 40% of whole consumption of raw coal in residential sector (79.07 Tg) (refer to 61).

pathways. One category is derived from free PAHs trapped in original coals, and the other is formed through pyrolysis and pyrosynthesis during coal burning (56). A large amount of coal tar is produced during the devolatilization period in coal burning, and this tar is composed mainly of aromatic species released either directly into the flue gas or reformed to other PAHs through further thermal degradation (26). Under the relatively low temperatures during residential coal combustion, a higher PAHs yield is expected, especially from bituminous and sub-bituminous coal with high volatile content. The poor air-supply in residential burning conditions may be another reason for the high production of PAHs. Ledesma et al. (26) concluded that most of the PAH mass is produced when the oxygen ratio is under 30% during bituminous coal burning. This indicates that it may be possible to decrease emission factors by improving the aeration conditions in residential combustion.

Previous studies on the emission factors of PAHs from various residential coal combustors are generally similar to our measurements (30–32, 46, 57). As indicated in those studies, the rank of coal (maturity) is an important factor that affects PAH emissions from residential combustion. Anthracite burning emits PAHs with a mass 3 orders of magnitude lower than bituminous. The burning conditions also affect PAHs emissions. For example, Oros and Simoneit (46) sampled including the smoldering conditions and reported the values of 2 orders of magnitude higher than that of others. Furthermore, large-scale combustors such as power plants have very low PAH emission factors, which are in the same order or even lower than that from anthracite in residential combustion, although most of the power plant fuels are bituminous coals (58–60).

Primary Estimates of Carbonaceous Particle and PAH Emissions. Primary estimates about the emissions of BC,

OC, and PAHs from residential coal combustion using honeycomb coal briquettes can be made simply as shown in Table 5. The total consumption of raw coal in the residential sector of China was about 79 Tg (Teragrams) during 2000 (3), in which 40% (31.6 Tg) was burned as honeycomb briquettes. Streets et al. (15) had assumed that briquettes would comprise 41% of domestic raw coal for the year 2020. Cheng (61) supposed that the briquette yield of China was 80 Mt during 2000. The mass of briquettes contained a large part of ash and clay, because the average ash content on air-dry basis of the five briquettes in the present work was about 45%. Using the percentages of anthracite (17.57%) and bituminous (78.07%, including sub-bituminous and bituminous) in the whole yield of raw coal (<http://www.cct.org.cn/cct/>), although more anthracite coal is expected to be utilized in large cities where raw coal is not available, there are 5.56 Tg of anthracite and 24.69 Tg of bituminous coal burned as honeycomb briquettes. So, the emissions of particulate matter, black and organic carbon, and PAH mass can be estimated, and the results are 290.24 Gg (gigagrams) for PM, 5.36 Gg for BC, 170.33 Gg for OC, and 4.72 Gg for total PAHs. As compared to the BC emissions of 605.4 Gg from domestic coal combustion by Streets et al. (15), honeycomb coal briquettes contribute less than 1%. However, it should be stressed that the emission factors of BC, OC, and PAHs could be underestimated in some degree because they were obtained only for flaming conditions, while smoldering (choking condition) was excluded because of the difficulties of sampling. Furthermore, other residential burning conditions of coal (60 wt %) besides briquette, such as unprocessed raw coal, whose emission factors of carbonaceous particles and PAHs are thought to be higher, remain uncertain and need further urgent investigations before any accurate estimates for emissions from residential coal combustion in China can be made.

Acknowledgments

This work was funded by the Chinese National Natural Science Foundation (No. 4013010). We would like to thank Prof. Xinming Wang and Mr. Zhengyue Li for their help in designing the sampling equipment, and Dr. Daishe Wu, Jingkui Mi, and Bofu Ning for collecting raw coal samples. Thanks are also given to Mr. Tongshou Xiang and Huizhi Zhang for technical assistance with instrumental analyses, and to Prof. Bernd R.T. Simoneit for his great help in revising this paper.

Literature Cited

- Jacobson, M. Z. Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming. *J. Geophys. Res.* **2002**, *107*, 4410.
- Lighty, J. S.; Veranth, J. M.; Sarofim, A. F. Combustion aerosols: factors governing their size and composition and implications to human health. *J. Air Waste Manage. Assoc.* **2000**, *50*, 1565–1618.
- China Statistical Yearbook, 2003*; China Statistics Press: Beijing, 2004.
- Cooke, W. F.; Liousse, C.; Cachier, H.; Feichter, J. Construction of a $1^\circ \times 1^\circ$ fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model. *J. Geophys. Res.* **1999**, *104*, 22137–22162.
- Jacobson, M. Z. Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. *Nature* **2001**, *409*, 695–697.
- Andreae, M. O. The dark side of aerosols. *Nature* **2001**, *409*, 671–672.
- Menon, S.; Hansen, J.; Nazarenko, L.; Luo, Y. F. Climate effects of black carbon aerosols in China and India. *Science* **2002**, *297*, 2250–2253.
- Qiu, J.; Yang, L. Variation characteristics of atmospheric aerosol optical depths and visibility in North China during 1980–1994. *Atmos. Environ.* **2000**, *34*, 603–609.
- Chameides, W. L.; Yu, H.; Liu, S. C.; Bergin, M.; Zhou, X.; Mearns, L.; Wang, G.; Kiang, C. S.; Saylor, R. D.; Luo, C.; Huang, Y.; Steiner, A.; Giorgi, F. Case study of the effects of atmospheric aerosols and regional haze on agriculture: an opportunity to enhance crop yields in China through emission controls? *Proc. Natl. Acad. Sci. U.S.A.* **1999**, *96*, 13626–13633.
- Kunzli, N.; Kaiser, R.; Medina, S.; Studnicka, M.; Chanel, O.; Filliger, P.; Herry, M.; Horak, F.; Puybonnieux-Texier, V.; Quenel, P.; Schneider, J.; Seethaler, R.; Vergnaud, J. C.; Sommer, H. Public-health impact of outdoor and traffic-related air pollution: a European assessment. *Lancet* **2000**, *356*, 795–801.
- Hamilton, R. S.; Mansfield, T. A. Airborne particulate elemental carbon: its sources, transport and contribution to dark smoke and soiling. *Atmos. Environ.* **1991**, *25*, 715–723.
- Ghan, S. J.; Chuang, C. C.; Easter, R. C.; Penner, J. E. A parametrization of cloud droplet nucleation, Part II: Multiple aerosol types. *Atmos. Res.* **1995**, *36*, 39–54.
- USEPA. Integrated Risk Information System (IRIS) on Polycyclic Organic Matter. National Center for Environmental Assessment, Office of Research and Development, Washington, DC, 1999; <http://www.epa.gov/ttn/atw/hlthef/polycycl.html>.
- Durant, J. L.; Lafleur, A. L.; Busby, W. F., Jr.; Donhoffner, L. L.; Penman, B. W.; Crespi, C. L. Mutagenicity of C₂₄H₁₄ PAH in Human Cells Expressing CYP1A1. *Mutat. Res.* **1999**, *446*, 1–14.
- Streets, D. G.; Gupta, S.; Waldhoff, S. T.; Wang, M. Q.; Bond, T. C.; Bo, Y. Y. Black carbon emissions in China. *Atmos. Environ.* **2001**, *35*, 4281–4296.
- Streets, D. G.; Bond, T. C.; Carmichael, G. R.; Fernandes, S. D.; Fu, Q.; He, D.; Klimont, Z.; Nelson, S. M.; Tsai, N. Y.; Wang, M. Q.; Woo, J.-H.; Yarber, K. F. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res.* **2003**, *108*, 8809.
- Cooke, W. F.; Wilson, J. J. N. A global black carbon aerosol model. *J. Geophys. Res.* **1996**, *101*, 19395–19409.
- Liousse, C.; Penner, J. E.; Chuang, C.; Walton, J. J.; Eddleman, H.; Cachier, H. A global three-dimensional model study of carbonaceous aerosols. *J. Geophys. Res.* **1996**, *101*, 19411–19432.
- Bond, T. C.; Covert, D. S.; Kramlich, J. C.; Larson, T. V.; Charlson, R. J. Primary particle emissions from residential coal burning: optical properties and size distributions. *J. Geophys. Res.* **2002**, *107*, 8347.
- Florig, H. K. China's air pollution risks. *Environ. Sci. Technol.* **1997**, *31*, 275A–279A.
- Chen, Y. J.; Bi, X. H.; Mai, B. X.; Sheng, G. Y.; Fu, J. M. Emission characterization of particulate/gaseous phases and size association for polycyclic aromatic hydrocarbons from residential coal combustion. *Fuel* **2004**, *83*, 781–790.
- Bi, X. H.; Sheng, G. Y.; Peng, P. A.; Chen, Y. J.; Zhang, Z. Q.; Fu, J. M. Distribution of particulate- and vapor-phase *n*-alkanes and polycyclic aromatic hydrocarbons in urban atmosphere of Guangzhou, China. *Atmos. Environ.* **2003**, *37*, 289–298.
- Birch, M. E.; Cary, R. A. Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust. *Aerosol Sci. Technol.* **1996**, *25*, 221–241.
- Butcher, S. S.; Ellenbecker, M. J. Particulate emission factors for small wood and coal stoves. *J. Air Pollut. Control Assoc.* **1982**, *32*, 380–384.
- Solomon, P. R.; Hamblen, D. G.; Carangelo, R. M.; Serio, M. A.; Deshpande, G. V. General model of coal devolatilization. *Energy Fuels* **1988**, *2*, 405–422.
- Ledesma, E. B.; Kalish, M. A.; Nelson, P. F.; Wornat, M. J.; Mackie, J. C. Formation and fate of PAH during the pyrolysis and fuel-rich combustion of coal primary tar. *Fuel* **2000**, *79*, 1801–1814.
- Mitra, A.; Sarofim, A. F.; Bar-Ziv, E. The influence of coal type on the evolution of polycyclic aromatic hydrocarbons during coal devolatilization. *Aerosol Sci. Technol.* **1987**, *6*, 261–271.
- Radke, M.; Schaefer, R. G.; Leythaeuser, D.; Teichmüller, M. Composition of soluble organic matter in coals: relation to rank and liptinite fluorescence. *Geochim. Cosmochim. Acta* **1980**, *44*, 1787–1800.
- Zhang, J.; Smith, K. R.; Ma, Y.; Ye, S.; Jiang, F.; Qi, W.; Liu, P.; Khalil, M. A. K.; Rasmussen, R. A.; Thorneloe, S. A. Greenhouse gases and other airborne pollutants from household stoves in China: a database for emission factors. *Atmos. Environ.* **2000**, *34*, 4537–4549.
- Oanh, N. T. K.; Reutergardh, L. B.; Dung, N. T. Emission of polycyclic aromatic hydrocarbons and particulate matter from domestic combustion of selected fuels. *Environ. Sci. Technol.* **1999**, *33*, 2703–2709.
- U.S. Environmental Protection Agency. *Compilation of Air Pollutant Emission Factors, AP-42*, 5th ed.; 1996; Vol. I, Chapter

- 1: External Combustion Sources, <http://www.epa.gov/ttn/chief/ap42/ch01>.
- (32) Mumford, J. L.; Harris, D. B.; Williams, K.; Chuang, J. C.; Cooke, M. Indoor air sampling and mutagenicity studies of emissions from unvented coal combustion. *Environ. Sci. Technol.* **1987**, *21*, 308–311.
- (33) Hangebrauck, R. P.; Lehmden, D. J. V.; Meeker, J. E. Emissions of polynuclear hydrocarbons and other pollutants from heat-generation and incineration processes. *J. Air Pollut. Control Assoc.* **1964**, *14*, 267–278.
- (34) Edwards, R. D.; Smith, K. R.; Zhang, J.; Ma, Y. Implications of changes in household stoves and fuel use in China. *Energy Policy* **2004**, *32*, 395–411.
- (35) Zhang, J.; Smith, K. R. Hydrocarbon emissions and health risks from cookstoves in developing countries. *J. Expos. Anal. Environ. Epidemiol.* **1996**, *6*, 147–161.
- (36) Kavouras, I. G.; Stephanou, E. G. Particle size distribution of organic primary and secondary aerosol constituents in urban, background marine, and forest atmosphere. *J. Geophys. Res.* **2002**, *107*.
- (37) Markowski, G. R.; Ensor, D. S.; Hooper, R. G. A submicron aerosol mode in flue gas from a pulverized coal utility boiler. *Environ. Sci. Technol.* **1980**, *14*, 1400–1402.
- (38) McElroy, M. W.; Carr, R. C.; Ensor, D. S.; Markowski, G. R. Size distribution of fine particles from coal combustion. *Science* **1982**, *215*, 13–19.
- (39) Dockey, D. W.; Pope, C. A.; Xu, X.; Spengler, J. D.; Ware, J. H.; Fay, M. E.; Ferris, B. G.; Speizer, F. E. An association between air pollution and mortality in six U.S. cities. *N. Engl. J. Med.* **1993**, *329*, 1753–1759.
- (40) Robinson, A. L.; Pandis, S. N.; Lipsky, E.; Stanier, C. Sampling, analysis, and properties of primary PM_{2.5}: application to coal-fired utility boilers. Annual Technical Progress Report, August, 2001.
- (41) Chow, J. C.; Watson, J. G. Recent developments in source characterization, presented at the regional particulate matter workshop, May 15–17, Hong Kong and Guangzhou, People's Republic of China, 2002.
- (42) Turpin, B. J.; Huntzicker, J. J.; Adams, K. M. Intercomparison of photoacoustic and thermal-optical methods for the measurement of atmospheric elemental carbon. *Atmos. Environ.* **1990**, *24A*, 1831–1835.
- (43) Mumford, J. L.; He, X. Z.; Chapman, R. S.; Cao, S. R.; Harris, D. B.; Li, X. M.; Xian, Y. L.; Jiang, W. Z.; Xu, C. W.; Chuang, J. C.; Wilson, W. E.; Cooke, M. Lung cancer and indoor air pollution in Xuan Wei, China. *Science* **1987**, *235*, 217–220.
- (44) Chung, J. C.; Wise, S. A.; Cao, S. R.; Mumford, J. L. Chemical characterization of mutagenic fractions of particles from indoor coal combustion: a study of lung cancer in Xuan Wei, China. *Environ. Sci. Technol.* **1992**, *26*, 999–1004.
- (45) Richter, H.; Howard, J. B. Formation of polycyclic aromatic hydrocarbons and their growth to soot – a review of chemical reaction pathways. *Prog. Energy Combust. Sci.* **2000**, *26*, 565–608.
- (46) Oros, D. R.; Simoneit, B. R. T. Identification and emission rates of molecular tracers in coal smoke particulate matter. *Fuel* **2000**, *79*, 515–536.
- (47) Bond, T. C. Light absorption by primary particles from fossil-fuel combustion: implications for radiative forcing. Ph.D. Dissertation, University of Washington, 2000; p 383.
- (48) Penner, J. E.; Eddleman, H.; Novakov, T. Towards the development of a global inventory for black carbon emissions. *Atmos. Environ.* **1993**, *27A*, 1277–1295.
- (49) Parungo, F.; Nagamoto, C.; Zhou, M. Y.; Hansen, A. D. A.; Harris, J. Aeolian transport of aerosol black carbon from China to the ocean. *Atmos. Environ.* **1994**, *28*, 3251–3260.
- (50) Chow, J. C.; Watson, J. G.; Pritchett, L. C.; Pierson, W. R.; Frazier, C. A.; Purcell, P. G. The DRI thermal/optical reflectance carbon analysis system: description, evaluation and applications in U. S. air quality studies. *Atmos. Environ.* **1993**, *27A*, 1185–1201.
- (51) Chow, J. C.; Watson, J. G.; Crow, D.; Lowenthal, D. H.; Merrifield, T. Comparison of IMPROVE and NIOSH carbon measurements. *Aerosol Sci. Technol.* **2001**, *34*, 23–34.
- (52) Schauer, J. J.; Mader, B. T.; Deminter, J. T.; Heidemann, G.; Bae, M. S.; Seinfeld, J. H.; Flagan, R. C.; Cary, R. A.; Smith, D.; Huebert, B. J.; Bertram, T.; Howell, S.; Kline, J. T.; Quinn, P.; Bates, T.; Turpin, B.; Lim, H. J.; Yu, J. Z.; Yang, H.; Keywood, M. D. ACE-Asia intercomparison of a thermal-optical method for the determination of particle-phase organic and elemental carbon. *Environ. Sci. Technol.* **2003**, *37*, 993–1001.
- (53) Yassaa, N.; Meklati, B. Y.; Cecinato, A.; Marino, F. Organic aerosols in urban and waste landfill of Algiers metropolitan area: occurrence and source. *Environ. Sci. Technol.* **2001**, *35*, 306–311.
- (54) Yassaa, N.; Meklati, B. Y.; Cecinato, A.; Marino, F. Particulate *n*-alkanes, *n*-alkanoic acids and polycyclic aromatic hydrocarbons in the atmosphere of Algiers City Area. *Atmos. Environ.* **2001**, *35*, 1843–1851.
- (55) Nisbet, C.; LaGoy, P. Toxic Equivalency Factors (TEFs) for Polycyclic Aromatic Hydrocarbons (PAHs). *Regul. Toxicol. Pharmacol.* **1992**, *16*, 290–300.
- (56) Mastral, A. M.; Callen, M. S. A review on polycyclic aromatic hydrocarbon (PAH) emission from energy generation. *Environ. Sci. Technol.* **2000**, *34*, 3051–3057.
- (57) Johnson, N. D.; Scholtz, M. T.; Cassaday, V.; Davidson, K. MOE Toxic Chemical Emission Inventory for Ontario and Eastern North America. Prepared for the Air Resources Branch, Ontario Ministry of the Environment, Rexdale, Ontario. Draft Report No. P.89-50-5429/OG, 1990; p 146.
- (58) Levendis, Y. A.; Atal, A.; Carlson, J. B. On the correlation of CO and PAH emissions from the combustion of pulverized coal and waste tires. *Environ. Sci. Technol.* **1998**, *32*, 3767–3777.
- (59) Yang, H. H.; Lee, W. J.; Chen, S. J.; Lai, S. O. PAH emission from various industrial stacks. *J. Hazard. Mater.* **1998**, *60*, 159–174.
- (60) Masclet, P.; Bresson, M. A.; Mouvier, G. Polycyclic aromatic hydrocarbons emitted by power stations, and influence of combustion conditions. *Fuel* **1987**, *66*, 556–562.
- (61) Cheng, Y. Q. On spreading the application of clean coal technology in China. *China Coal* **1998**, *24*, 12–16.

Received for review April 26, 2004. Revised manuscript received December 13, 2004. Accepted December 14, 2004.

ES0493650