Emission of Polycyclic Aromatic Hydrocarbons from Indoor Straw Burning and Emission Inventory Updating in China

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The emission factors for indoor straw combustion are a major data gap for estimating the polycyclic aromatic hydrocarbon (PAH) emissions in China. The emission factors for open-fire straw burning were borrowed from our previous study and a rough estimate was developed. As one of the most important emission sources in China, the emission factors for indoor straw combustion needed to be determined and revised accurately. In this study, a representative straw in rural China was collected and burned in similar conditions with those used by countryside families. The smoke produced was sampled and the PAH concentrations were analyzed by gas chromatography-mass selective detection (GC-MSD), and much higher emission factors were found. Based on the newly measured emission factors, the emission amount from indoor straw combustion was updated. In addition, recently published emission factors were compiled in a comprehensive database and some new sources were included. Additionally, the emission inventory was extended to cover the period from 1950 to 2005 and upgraded to a scale resolution of one kilometer. In the updated inventory, the total quantity of 16 PAHs emitted from China was 116,000 tons in 2003, with indoor straw and firewood combustions as the most important sources. Although vehicular emission contributed a relatively small percentage of the total emission, it was still one of the major sources in the urban areas of China. The total PAH emission increased continuously for four decades, starting from 1950, but fluctuated since 1990 due to variations in coke production.

Key words: PAHs; emission factor; indoor straw burning; emission inventory

Introduction

In a previous study, an emission inventory for polycyclic aromatic hydrocarbons (PAHs) in China was developed. Based on emission activity strengths and emission factors collected from the literature, the annual emission of 16 USEPA priority PAHs in 2003 was estimated to be 25,000 tons.¹ The major emission sources were identified as straw and firewood burning, industrial and domestic coal combustion, coke production, transport and nontransport petroleum combustion, and primary aluminum production.¹ However, uncertainty in the estimation was addressed, and it was indicated that the lack of accurate emission factors for PAHs, particularly those measured at local conditions, was the primary source of the ambiguity. A typical example was that PAH emissions from straw burning were pooled together due to a data gap on emission factors for indoor straw burning.

In China, straw is usually burned in two ways: open burning as agricultural waste in fields, and indoor burning as fuel in cook stoves.² The latter is an important energy source in rural China. Approximately 340 million tons of straw was consumed for cooking and indoor heating by conventional household

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stoves in China in 2004.³ This value was about 60% higher than that of the residential firewood combustion, which was usually believed to be the predominant biofuel used.³ In fact, emission of PAHs from indoor firewood combustion was extensively investigated.^{4–6} However, except for a study of the PAH emission profile for the PM2.5,⁷ the total PAH emission factors, including both particulate and gaseous phases from indoor straw burning, had never been reported.

The emission of PAHs from carbonaceous fuel combustion depends largely on the combustion conditions and the properties of different fuels.⁸ Zhang et al. also reported very different emission factors of PAHs in PM2.5 for several types of straw between smoldering and flaming conditions.⁷ For the purpose of more accurate evaluation, it is desirable if the different combustion conditions can be distinguished. However, the emission factors reported for straw burning, to date, were all responsible for open-fire burning of straw as agricultural waste in the fields, and the straw burned under indoor and open-fire conditions were not distinguished in our previous study.¹ Recognizing the fact that indoor straw combustion is one of the major PAH emission activities, derivation of firsthand PAH emission factors from indoor straw burning in China would be critical for the updating of the PAH emission inventory for China. In addition to straw burning, other activities, including small-scale coke ovens, also introduced uncertainty into the inventory due to the limitation in emission factors.

The objective of this study was twofold: (1) to measure PAH emission factors of indoor straw burning, and (2) to update the PAH emission inventory in China. Instead of county resolution estimation from 1980 to 2003,⁹ the revised inventory was developed based on a $1 \times 1 \text{ km}^2$ grid and extended to a period from 1950 to 2005. The 16 parent PAHs included in this study were: naphthalene (NAP), acenaphthylene (ACY), acenaphthene

(ACE), fluorene (FLO), phenanthrene (PHE), (ANT), fluoranthene (FLA), anthracene pyrene (PYR), benz(*a*)anthracene (BaA),benzo(*b*)fluoranthene chrysene (CHR),(BbF),benzo(*k*)fluoranthene (BkF),benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DahA), indeno(l,2,3-cd)pyrene (IcdP), and benzo(g,h,i) pervlene (BghiP). The total 16 PAHs and the sum of the 7 carcinogenic PAHs, namely, BaA, CHR, BbF, BkF, BaP, IcdP, and DahA, are denoted as PAH16 and PAH7, respectively.

Methodology

Determination of Emission Factors for Indoor Straw Burning

Burning and Sampling System

The straw sample in this study was collected from the neighboring areas of Beijing. With a large planting area and output in China, wheat was selected as the representative type of cereal straw. The straw was cut into small sections with a length of about 5 cm for each, and piled up for combustion on a metal plate. The burning and sampling system is illustrated in Figure 1. The measuring experiment was finished in an independent laboratory room to avoid interference from possible organic contamination. The sampling device was mainly composed of a hood, a dilution tunnel, and a blower. The hood was made of smooth sheet iron, with a height of 1.2 m, and the bottom was a square of $1.5 \times 1.5 \text{ m}^2$. The dilution tunnel, made of stainless iron, was about 3.5 m long, with a diameter of 0.127 m, in which the combustion fume was mixed with clean ambient air introduced by the blower and cooled. It should be indicated that the inside tunnel length of turbidity was longer than 2 m, greater than 10 times that of the inner diameter of the dilution tunnel (0.127 m), which can ensure full mixing. At the end of the dilution tunnel, a high-flow rate blower was mounted for pumping during the burning procedure. An S-type pitot tube



Figure 1. Setup of the facilities for measuring polyclinic aromatic hydrocarbons (PAH) emission factors from indoor straw combustion. The straw is burned on a plate and a balance is continuously measuring the fuel burn rate. The hood and the blower directed the yielded smoke into a pipe, where the smoke was mixed with ambient air by a turbulent current. The PAH sampler (consisting of a prepositioned glass fiber filter (GFF) and two polyurethane foam filters (PUFs) in tandem) collected both the gaseous and particulate phases of the combustion gas stream from this pipe.

was utilized to measure the average velocity on the cross section in the tunnel.

Based on the preliminary experiments, the inner dimension of the dilution tunnel was sufficient for full mixing, dilution, and cooling, at the current velocity of 10.0 L/min. To maintain a constant dilution ratio, the blower voltage (U) was adjusted to control the inlet volume of clean air. When the U was set at 180 V as the normal working voltage, the flow rate stably reached 17.5 m/s, and the dilution ratio was suitable for sampling. The objective compounds in produced smoke were captured by a combined active sampling device installed at the end of the tunnel, as shown in Figure 1. The sampling device was driven by an electronic pump (TMP 1500; Jiangsu Eltong Electric Corporation, Co., Ltd., China). To avoid the reduction in adsorption efficiency of polyurethane foam (PUF; 22 mm o.d. \times 7.6 cm length) due to the high concentration of smoke, an assembled cartridge having two glass PUF disks in tandem and a prepositioned glass fiber filter (GFF; diameter = 80 mm) were employed

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to collect the gaseous and particulate-phase PAHs, respectively. The pumps were calibrated with a flow rate of around 0.93 L/min, and the sampling time was ascertained based on straw combustion conditions, normally for 1.5 h or so. Samples for fuel gas and samples for ambient air in the laboratory room were collected simultaneously as the controls. During the whole combustion procedure, the weight of the straw sample was recorded every 5 min to monitor the combustion process. The smoke yielded was collected by a rectangular hood and pumped through a pipe where the smoke and the ambient air introduced were mixed thoroughly by a turbulent current. After collection, the PUF disks and GFFs were stored at -18° C until the analysis was performed.

Extraction, Purification, and Analysis

The PUF disk and GFFs were extracted by Soxhlet in a 1:1 mixture of *n*-hexane and cyclohexane over a period of 4 and 10 h, respectively. The extracts were concentrated to about 1 mL by a rotary evaporator, and calibrated to 1 mL with n-hexane in a vial. The samples were analyzed by gas chromatograph that was equipped with a mass selective detector (MSD) (Agilent 6890N/5973I, 30 m length $\times 0.25 \text{ mm}$ i.d. \times 0.25 µm film thickness, HP-5 MS capillary column; Agilent Technologies Irvine, CA) with helium as the carrier gas. The injection volume was 1 µL in splitless mode. The column temperature was programmed from 60°C to 300°C at 5°C/min, and then kept constant for 15 min. The MSD was operated in electronimpact mode at 70 eV with an ion source temperature of 230°C. Quantification was performed by the external standard method using the mixed external standard of the 16 PAHs (U.S. Chemical Service; Chem Service Inc., West Chester, PA).

QA/QC

The PUF disk blank and the GFF were measured by the same procedure and subtracted from the tested samples. A sample without straw burning was also measured to serve as the procedure blank and was subtracted as well. Except for NAP in the GFF, all the blanks were more than one order of magnitude lower than the sample measurement. The preliminary experiments indicated that the PAH concentrations in the second PUF disks were about two orders of magnitude lower than the first one, except for NAP (only about three times lower), which guaranteed that nearly all the gaseous PAHs were collected by this combined sampler. All the measured results were corrected by the method recoveries, and ranged from 70% to 142% for the 16 individual PAHs.

Updating PAHs Emission Inventory in China

To update the PAH emission inventory, the emissions from open-field straw burning and indoor straw combustion were calculated individually, and the emission factors derived in our laboratory were applied to the latter. In the meantime, the emission factors for indoor firewood combustion and coke production were recompiled by including recently reported data. For instance, only the emission factor of BaP from the small-scale coke ovens was included.¹⁰ In order to give a rough estimation of the PAH emission from this source, the emission factors reported for uncontrolled coke production were adopted, based on an assumption that the emission control conditions of small-scale coke production in China are similar to those of the so-called "beehive" coke ovens in the United States in 1900s.^{11,12} This assumption was partially confirmed by the similarity between the calculated and the reported BaP emission factors.^{10,11} Emission factors for newly added emission activities, including petroleum refinery, gasoline distribution, and consumer product usage, were also compiled based on the data from the literature.

The quantity of straw burning in a field, small-scale coke produced by crude ovens, refined petroleum, and gasoline distributed in China were derived from the literature. The use of consumer goods, including personalcare products, household products, automobile after-market products, and other commercial and consumer products that may emit PAHs during usage, were considered as a single source and calculated by multiplying population and per capita emission, which was assumed to be proportional to the per capita household consumption expenditure.

Physical and socioeconomic data at $1 \times 1 \text{ km}^2$ resolution were collected from the Chinese Natural Resources, Environment, Economic and Population Database (available at: http://www.naturalresources.csdb.cn, in Chinese). The regression models between the emission activity strength and these parameters developed by Zhang et al. were further modified and adopted to predict PAH emission activities at $1 \times 1 \text{ km}^2$ resolution.⁹ Similar models were developed for the new sources identified in this study. It was also assumed that the indoor straw and firewood burning occurred only in rural areas, and that open-fire straw burning presented in agricultural land. All industrial sources were allocated to urban areas. For point sources like coke and aluminum production, the same method used in the mapping at the county level was adopted.⁹ ARCGIS 9.0 was employed for data storage and manipulation (ESRI, Inc., Redlands, CA).

Results and Discussion

Emission Factors for Indoor Straw Burning

The measured emission factors for each individual PAH from indoor straw burning are shown in Table 1. As most PAH compounds studied are semivolatile and can distribute between the gaseous and particulate phases, the emission factors corresponding to the two phases are both shown. The total emission factor for PAH16 in residential wheat-straw combustion was about 343 mg/kg, with about 60% distributed in the gaseous phase, while

rable 1.	Tested Poly	clinic Aro	matic H	ydrocark	on Emis	sion Fac	tors (mg	ı/kg) for	Indoor \$	itraw Bu	ning-						
Phases	NAP	ACY	ACE	FLO	PHE	ANT	FLA	PYR	BaA	CHR	BbF	BkF	BaP	IcdP	DahA	BghiP	PAH16
Gaseous	108.8	31.7	10.6	18.8	18.0	6.3	2.3	1.8	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	198.4
Particulate	0.1	1.5	0.3	3.8	22.7	8.9	24.1	24.4	15.8	17.8	5.4	7.8	6.6	2.5	0.4	2.6	144.8
Total	108.9	33.2	11.0	22.6	40.7	15.2	26.4	26.3	15.8	17.9	5.4	7.8	6.6	2.5	0.4	2.6	343.3

in Table 2. Except for NAP, the emission factors corresponding to the other 15 compounds for indoor straw combustion were much higher than those for open-fire straw burning, especially for the low molecular weight species from ACY to CHR, with about one order of magnitude. The differences in emission factors between indoor and open-fire straw combustion can be largely caused by the different amount of oxygen supply. Jenkins et al. developed a windtunnel system to simulate the spreading fires during the burning of agricultural wastes in fields by farmers.¹³ In order to better simulate the combustion environment of wild fires, ambient air was offered to the burning straw by a blower.¹³ This setting provided a sufficient oxygen supply for the straw combustion. On the other hand, in an indoor combustion environment, the air flow was largely limited by the indoor environment, which resulted in an oxygen-deficient atmosphere. The pyrolysis of the fuel material to free radicals and the pyrosynthesis of these radicals are usually considered as the two main steps during the formation of PAHs. Free radicals are first produced by the breaking of the carbon-carbon and carbon-hydrogen bonds in fuels at high temperature. Together with the unburned fuel materials, the radicals can undergo fast condensation and association reactions to further form aromatic ring structures. However, oxygen can compete with the pyrosynthesis process by oxidizing the free radicals, which means that more PAHs will be formed in the oxygen-deficient atmosphere.⁸ Other factors, like burning rate and temperature, may also contribute to the difference in emission factors, but they will not be discussed here due to limited size of the available data.

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in the case of PAH7, it was 56 mg/kg, which accounted for around 16% of the total emission and was mainly distributed in the particulate phase.

The measured emission factors for indoor straw combustion were compared with those for open-fire straw burning in a field, as shown

TABLE	2. Comparisor	ו between the א	Aeasured	and th	ie Litei	rature	-Repor	ted Pc	olycycl	ic Aroı	natic	Hydro	carbo	n Emi	ssion F	actors .	for Stra	w Burnir	ō
Straw	Configurationre	p Burning type	NAP	ACY	ACE	FLO	PHE	ANT	FLA	PYR	BaA (CHR	BbF F	kF B	aP Icd	P DahA	BghiP	PAH16	Ref
Wheat ^a	CEWF ^a 1	Open burning	44.7	0.3	0.7	0.4	5.7	1.3	7.3	4.5	2.4	2.5	2.9	0.8	.0 0.	1.2	1.1	77	13
Wheat	CEWF 2	Open burning	44.1	0.4	1.1	0.2	4.8	1.2	6.0	3.3	2.2	2.1	1.0	0.5 0	.3 0.(0.2	1.0	68	13
Wheat	CRNF ^b 1	Open burning	669.3	0.0	3.6	0.6	3.4	1.1	1.3	1.1	0.3	0.4	0.5	0.3 0	.2 0.0	0.0	0.0	682	13
Wheat	CRNF 2	Open burning	26.7	0.0	0.6	0.1	2.6	0.7	I.I	1.0	0.4	0.4	0.1	0.3 0	.1 0.0	0.0	0.0	34	13
Wheat		Indoor burning	108.9	33.2	11.0	22.6	40.7 I	5.2	26.4 2	26.3 1	5.8	17.9	5.4	7.8 6	.6 2.5	0.4	2.6	343.3	this
																			study

CEWF = ceiling extended with floor; both the floor and ceiling were employed.

CRNF = ceiling retracted, no floor; neither the floor nor the ceiling was employed.

Emission Amount from Indoor Straw Combustion

According to the newly determined emission factors in this study, the PAH16 emission from indoor straw combustion was estimated to be 43,700 tons, which is much higher than the previous estimation of 6500 tons.¹ The significant increase resulted in the emission from indoor straw burning exceeding that from residential firewood combustion (24,000 tons), and the former became the largest emission source for PAHs in China.

Besides the relatively high emission factors, a large quantity of straw was widely consumed by the rural residents for cooking or heating in the residential sector and caused high emission amounts. In China, about 620 million tons of straw was produced in 2002, and it was estimated that 45% of the total straw yield was used as fuel in China.² Generally, about 33-45% of the residential energy needed in rural China was supplied by straw combustion. Furthermore, if we count the biofuel only, straw would contribute about 60% to the total amount of biofuel consumed.³ Although other traditional materials consumed, such as forage and building materials, have been gradually replaced by commercial products as the income per capita increases in rural China, the guantity of straw used as fuel has kept increasing in recent years due to its extremely low cost and ready availability.² As a result, 370 million tons of straw, compared with 340 million and 330 million tons in 2004 and 2003, respectively, were burned indoors for cooking and heating in 2005.³ The combustion was usually performed in conventional stoves with poor ventilation, and the emission of PAHs is largely enhanced due to lack of oxygen supply.⁸ Only a small portion of straw was utilized by some other, cleaner techniques, like anaerobic digestion and straw gasification.¹⁴ Considering anaerobic digestion as an example, this technique is the most widely used in rural China. There were 7.64 million household biogas digesters in China in 2000, and about 22.74 billion m³ of biogas were



Figure 2. Relative contributions of various sources to the total emission of PAH16 in China in 2003.

produced.² However, the energy fraction originating from this technique only accounted for less than 2.5% of the total energy from straw.^{2,3}

Emission Inventory Updating

The PAH emission inventory in China was updated by revising the emission activity and emission factor database based on the recently measured and published data. The emissions of PAH16 and PAH7 emitted in China in 2003 were revalued as 116,000 tons and 11,800 tons, respectively. The differences from the previous estimations were largely caused by the updating of the emission factors. Besides the increase of emission from indoor straw burning, emission from coke production also increased from 4100 tons to 33,000 tons in 2003 as a result of calculating the emissions for the small-scale and industrial-scale ovens individually. Differences were also caused by subcategorizing individual activities such as vehicle and straw types. Newly included sources, particularly consumer and commercial product usage, contributed to the increase with a total emission of about 2000 tons.

The updated relative contributions of various sources to PAH emission are presented in Figure 2. Indoor straw combustion (34.6%), the small-scale coke production (27.2%), firewood burning for cooking and heating (21.2%), and domestic coal combustion (6.8%) dominated the total emission. Other emission sources added up to merely about 10% of the total. Emission from motor vehicles accounted for only 2.5% of the total, due to relatively low emission factors. Although vehicular emission contributed to a small percentage of PAH emission in China as a whole, they were among the major sources in Chinese cities, resulting in a mean emission density in urban areas 30 times higher than that in rural China. As such, it was often reported in the literature that traffic is the major PAH emission source in urban areas in China.^{15–17}

Geographical Distribution

In addition to the regression models developed in our previous study, similar models were developed for the ratios of straw burned in open fires to the total straw yielded, refined petroleum, and distributed gasoline as functions of per capita income in rural areas, secondary, and tertiary gross domestic products (GDPs), respectively. All three activities were well predicted by the simple linear models with coefficients of determination (r^2) from 0.70 to 0.82. Using these models and the physical and socioeconomic parameters collected in the literature, the emission activity strengths were predicted based on a regular $1 \times 1 \text{ km}^2$ grid system, which allows the emissions to be calculated by being multiplied by the emission factors.

The emission density map is presented in Figure 3. For most provinces, an urbancentered high emission density pattern is demonstrated. A few examples include Harbin, Changchun, and Shenyang in northeast China; Tianjin, Shijiazhuang, and Jinan in north China; the Pearl River Delta city cluster including Guangzhou and Hong Kong in south China; the Yangtze River Delta metropolitan area in east China; and Hohhot, Urumqi, and Lhasa in the remote provinces. In addition to these large cities or city clusters, many small-medial cities are also identified on the map. Such a correlation between PAH emission density and population density agrees with the relationship observed between atmospheric PAH concentration and local population density in North America and other developed



Figure 3. PAH16 emission-density map of China with spatial resolution of 1×1 km².



Figure 4. Changes in (**a**) the annual emission activities and (**b**) PAH16 emission in China from 1950 to 2005.

countries.¹⁸ Two exceptions were Shanxi and Guizhou provinces, where PAH emission from coke production dominated, both provinces being rich in coal reserves.^{19,20} Therefore, the geographical distribution of PAH emission density in the two provinces are primarily coal mine related.

Temporal Variation from 1950 to 2005 and Projection

The PAH emission inventories from 1950 to 2005 were developed and are presented in Figure 4 together with emission activities. Although several new emission sources were included, the general trend of temporal change was similar to that reported previously, except for a fluctuation since 1990. Starting from 1950, PAH emission increased gradually with the growth of total energy consumption in China. For the major PAH emission activities, the consumption of straw, firewood, domestically combusted coal, and coal for smallscale coke ovens increased from 87, 60, 41, and 0.3 million tons in 1950 to 265, 171, 244, and 30.7 million tons in 1990, respectively.³ Subsequently, PAH16 emission increased from 19,000 tons to 89,000 tons during the same period. Since the beginning of the 1990s, the annual emission of PAHs has fluctuated primarily due to the change in small-scale coke production driven by market demand and enforcement of the Coal Law of China in 1996.²¹ During the last couple of years, total energy consumption in China jumped dramatically, and the amount of industrially used coal increased from 950 million tons in 2000 to 1650 million tons in 2005, which corresponded with the fast growth pace of the GDP.³ However, with relatively low PAH emission factors,⁸ such an increase in industrial coal consumption had limited influence on PAH emission.

Based on the prediction of the International Energy Agency (the World Energy Outlook 2004, available at: http://www.iea.org), energy consumption in China will keep increasing for at least the two decades to come. It was predicted that the annual growth rates of coal and biomass consumption would be 2.3% and 0.3%, respectively, implying a scenario of slight increases for PAH emission. On the other hand, starting from 2004, much stricter regulation of small-scale coke production has been enforced, and over 40% of these facilities have been closed in China.²² The phasing out process will be accomplished by 2010, in accordance with the Coal Law.²¹ Since small-scale coke production contributed to a large share of the total PAH emission (18.5% in 2004), this measure, if implemented thoroughly, can substantially reduce the total PAH emission.

As the largest emission source, indoor straw combustion contributed to more than one-third of the total emission. Therefore, controlling the emissions from this source can efficiently reduce the PAH emission in China. The Chinese government launched the National Improved Stove Program in 1980, and by the end of the year 2000, about 189 million conventional household stoves had been substituted by improved ones, which accounted for about 80% of the total stoves in rural China.² These improved stoves have higher heat efficiency and can save about 1 ton of fuel per stove per year.¹⁴ However, the potential for further reducing the straw combustion by this approach may be limited for the reason that only a small number of conventional stoves remain now. Other ways of utilizing straw, including anaerobic digestion, straw gasification, straw briquette, straw liquefaction, straw carbonization, and biocoal, are

among the most possible ways for transferring straw to energy and reducing the amount of straw directly burned.² These techniques developed rapidly in recent years, and some of them have been widely promoted in rural China. For instance, the number of household biogas digesters has increased from about 4.7 million in 1990 to about 7.6 million in 2000, with an average annual increase rate of 4.8%.^{2,14} Similarly, 388 straw gasification facilities for central gas supply were also built by the end of the year 2000, and since then about 150 million m^3 of biogas have been provided each year.³ Currently, great efforts have been made to develop more affordable and reliable straw utilization techniques.² Although there are still technical, financial, as well as management difficulties in technique development and popularization, the potential for reducing PAH emission will be enormous.

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Conflicts of Interest

The authors declare no conflicts of interest.

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