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Greenhouse gases and other airborne pollutants from household stoves in China: a database for emission factors

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Abstract

Emissions from household stoves, especially those using solid fuels, can contribute significantly to greenhouse gas (GHG) inventories and have adverse health impacts. Few data are available on emissions from the numerous types of cookstoves used in developing countries. We have systematically measured emissions from 56 fuel/stove combinations in India and China, a large fraction of the combinations in use world-wide. A database was generated containing emission factors of direct and indirect GHGs and other airborne pollutants such as CO₂, CO, CH₄, TNMHC, N₂O, SO₂, NO_x, TSP, etc. In this paper, we report on the 28 fuel/stove combinations tested in China. Since fuel and stove parameters were measured simultaneously along with the emissions, the database allows construction of complete carbon balances and analyses of the trade-off of emissions per unit fuel mass and emissions per delivered energy. Results from the analyses show that the total emissions per unit delivered energy were substantially greater from burning the solid fuels than from burning the liquid or gaseous fuels, due to lower thermal and combustion efficiencies for solid-fuel/stove combinations. For a given biomass fuel type, increasing overall stove efficiency tends to increase emissions of products of incomplete combustion. Biomass fuels are typically burned with substantial production of non-CO₂ GHGs with greater radiative forcing, indicating that biomass fuels have the potential to produce net global warming commitments even when grown renewably. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

It is estimated that biomass combustion contributes 20–50% of global greenhouse gas (GHG) emissions

(Crutzen and Andreae, 1990; IPCC 1990). In recent years, the wide uncertainty about the emissions from open large-scale biomass fires has led to a substantial increase in attention given to measuring emissions from wildland fires (Ward and Hardy, 1991), prescribed fires (Cofer et al., 1993), and agricultural burning (Jenkins et al., 1992). A significant fraction of all biomass combustion, however, occurs in enclosed or semi-enclosed small combustion devices such as household stoves for cooking and space heating. For example, about 90% of the firewood

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(FAO, 1987) and a large fraction of the combusted crop residues are burned in household stoves worldwide, mainly in developing countries (Smith et al., 1993, 1999; Wang, 1994).

Household stoves, although individually small, are numerous and thus have the potential to contribute significantly to inventories of greenhouse gases (GHG), particularly in those many developing countries where household use is a significant fraction of total fuel use. The emissions of greenhouse gases from small-scale combustion of biomass are not well characterized (IPCC, 1997), but are known to be different from open large-scale combustion, such as forest and savannah burning (Levine, 1996). Unfortunately, few measurements have been made to determine emission factors for biomass stoves in developing countries (Smith et al., 1993). Emission factors from other fuels (e.g., coal, kerosene) as commonly used in developing-country households are also not well characterized. Therefore, measurements of GHG emission factors from a range of fuels and combustion devices would remove some of the uncertainty in estimates of global GHG emissions from fuel combustion and also provide a baseline for understanding the potential for reduction in GHG emissions due to various mitigation measures, such as fuel switching, in the household sector.

A pilot study was conducted in Manila, the Philippines, to measure emission factors of a range of GHGs from cookstoves burning biomass, charcoal, kerosene and liquefied petroleum gas (LPG). This pilot study validated a carbon balance approach for determining emission factors for small-scale combustion devices and resulted in tentative, but important, findings regarding carbon dioxide (CO₂) and non-CO₂ GHG emissions from simple household stoves (Smith et al., 1993). To explore these tentative findings further and to produce a comprehensive database of GHG emission factors for a wide range of household stoves commonly used in developing countries, we have tested a total of 56 fuel/stove combinations in India and China for a range of GHGs and other emissions while simultaneously monitoring for fuel and stove parameters. The chosen fuel/stove combinations represent a large fraction of the total in developing countries. The results derived from 28 Indian fuel/stove combinations, along with the details of the methods, have been presented previously (Smith et al., 1999). In this paper, we present a database of emission factors of the following species for the 28 fuel/stove combinations tested in China: CO₂, carbon monoxide (CO), methane (CH₄), total non-methane hydrocarbons (TNMHC), total suspended particles (TSP), and carbon fraction in TSP (C_{TSP}), nitrogen oxide (NO_x), and sulfur dioxide (SO₂).

2. Methods

2.1. Fuel/stove combination tested

The fuel types selected represent those being used commonly in rural households (e.g., crop residue, wood, kerosene, and coal) and those in urban households (e.g., gaseous fuels, coal, and kerosene). The stove types selected were those most typical for burning each type of fuels (i.e., most popular models found in the market or rural households). Since the emission factors are expected to depend upon both fuel types and stove types (Zhang and Smith, 1996), it is most appropriate to discuss the results by “fuel/stove combination”. Thus, the fuel/stove combination was treated as elementary unit for the tested household stoves. In total, 28 fuel/stove combinations were tested. Except those stoves using piped gas fuels, which were measured in actual homes, all other fuel/stove combinations were tested in a simulated village kitchen house at Tsinghua University’s rural campus about 50 km from Beijing.

The 28 fuel/stove combinations tested are shown in Table 1. Within each fuel category, a few fuel varieties were tested. Wood fuels and crop residues were obtained from a local market in a small town near Tsinghua University’s rural campus. Brush wood refers to thin branches of brushes which normally grow faster than trees. Brush wood is usually cheaper than fuel wood which is normally harvested from mature trees or their big branches (e.g., eucalyptus, acacia, oak, pine, poplar, willows, etc). Maize residue and wheat residue refer to dried corn stalks and wheat straws, respectively. Except unprocessed coal powder from a specific coal mine in Shanxi Province, all the other coals were obtained in local markets near Beijing. Washed coal was prepared by soaking unprocessed coal in clean water for 24 h, a typical practice in China to reduce sulfur content of raw coal. After washing, the coal powder was sun dried. Coal briquettes, or coal balls, are made of coal powder and clay. Honeycomb coal briquettes, also made of coal powder and clay, are named so due to their honeycomb-like shape. Elemental compositions of the tested solid fuels are shown in Table 2. All solid fuels were procured in one lot, sun-dried, and stored in a large storage room for at least 4 weeks prior to the tests.

Kerosene is a middle distillate from petroleum refining and is in liquid form under normal atmospheric conditions. Carbon, hydrogen, and oxygen content of the kerosene tested were 85.6, 13.0, and <0.5% (by weight), respectively. Liquid petroleum gas, (LPG), another product of petroleum refining, is available in bottles and is in gaseous form under normal atmospheric conditions. The tested LPG consisted of 26.7% propane, 18.8% butane, 43.4% butene, and 11.1% of other hydrocarbons (by volume). Coal gas is synthesized from coal gasification and distributed locally to households through pipelines.

Table 1
Description of the fuel/stove combinations tested in China

Serial #	Symbol fuel/stove	Description	
		Fuel	Stove
1	Brush-Brick-v	Brush wood	brick stove with a flue
2	Brush-Imp-v	Brush wood	improved brick stove with a flue
3	Brush-India	Brush wood	metal stove without flue (from India)
4	Wood-Brick-v	Fuel wood	same as # 1
5	Wood-Imp-v	Fuel wood	same as # 2
6	Wood-India	Fuel wood	same as # 3
7	Maize-Brick-v	Maize residue	same as # 1
8	Maize-Imp-v	Maize residue	same as # 2
9	Wheat-Brick-v	Wheat residue	same as # 1
10	Wheat-Imp-v	Wheat residue	same as # 2
11	CoalBriq-metal	Coal briquette	metal coal stove without flue
12	CoalBriq-metal-v	Coal briquette	metal coal stove with a flue
13	Honey-imp	Honeycomb coal briquette	improved metal coal stove without flue
14	Honey-metal	Honeycomb coal briquette	same as # 11
15	Honey-metal-v	Honeycomb coal briquette	same as # 12
16	Honey(s)-metal-v	Honeycomb coal from a special coal mine in Shanxi province	same as # 12
17	Coal-brick-v	Unprocessed coal (coal powder)	same as # 1
18	Coal-metal	Unprocessed coal (coal powder)	same as # 11
19	Coal-metal-v	Unprocessed coal (coal powder)	same as # 12
20	Coal(s)-metal-v	Coal powder from a special coal mine in Shanxi province	same as # 12
21	WashCoal-metal-v	Washed coal powder	same as # 12
22	Kero-press	Kerosene	kerosene pressure stove without flue
23	Kero-wick	Kerosene	kerosene wick stove without flue
24	LPG-IR	Liquefied petroleum gas	LPG stove with an infrared head without flue
25	LPG-trad	Liquefied petroleum gas	LPG traditional stove without flue
26	NaturalGas-IR	Natural gas	gas stove with an infrared head without flue
27	NaturalGas-trad	Natural gas	traditional gas stove without flue
28	CoalGas-trad	Coal gas	traditional gas stove without flue

Note: flue code: v = vented, i.e., with flue.

The lines in the table segregate the fuel category: # 1-6 wood; # 7-10, crop residue; # 11-21, coal; # 22-23, kerosene; and # 24-28, gas.

The tested coal gas consisted of 61.4% hydrogen, 23.8% CH₄, 5.6% CO, 2.7% CO₂, 3.7% nitrogen, 0.1% oxygen, and 2.7% NMHC (by volume). The natural gas tested in a Beijing home was transported from a natural gas field located more than 1000 km northwest of Beijing and consisted of 84.5% CH₄, 0.8% CO₂, 0.7% nitrogen, and 14% NMHC (by volume).

The brick and improved stoves in Table 1 were built on the floor with their flues attached to a side wall of the simulated kitchen (a typical arrangement found in village kitchens). The India stove was a portable metal non-chimney stove made in India and transported to China for the study. All the other stoves were purchased from local markets. Although similar to the (traditional) stoves in overall shape and structure, the improved stoves had a better design to increase stove thermal efficiencies (Gu et al., 1991). The kerosene wick stove tested was a portable, one-burner stove. This was the most popular type of

kerosene stove in China. The kerosene pressure stove, a single-burner pumping stove, is much less popular and more costly than the kerosene wick stove in China. All stoves used for burning gaseous fuels were standard multiple-burner gas ranges, although only one burner was used for the tests. The infrared head, a circular device attached around the burner under the pot is designed to convert a portion of the heat released into the surrounding air into infrared radiation which can then irradiates the pot bottom.

2.2. Carbon balance approach

The mass balance of carbon in fuel combustion process can be described as follows,

$$C_f - C_a = C_{CO_2} + C_{CO} + C_{CH_4} + C_{TNMHC} + C_{TSP}, \quad (1)$$

Table 2
Composition of solid fuels tested

Fuel	Carbon (%)	Hydrogen (%)	Sulfur (%)	Water (%)	Ash (%) ^a
Brush wood	44.48	5.39	0	7.65	1.75
Fuel wood	45.58	4.74	0	7.98	1.16
Maize residue	34.81	3.56	0	9.09	6.28
Wheat residue	40.34	5.29	0	7.26	8.71
Coal briquette	49.39	0.56	0.257	3.48	43.35
Honeycomb coal briquette	63.65	0.28	0.166	3.94	25.36
Honeycomb coal from a special coal mine in Shanxi province	54.33	2.6	0.94	2.68	35.8
Unprocessed coal powder	73.76	4.38	0.852	2.08	7.2
Coal powder from a special coal mine in Shanxi Province	62.55	2.66	0.98	1.00	29.87
Washed coal	77.75	4.52	0.35	4.73	3.7

^aAsh content of fuel, not the actual ash generated by the combustion.

where C_f is the carbon mass in the fuel, C_a the carbon mass in the ash including remaining unburned solid fuel, or char, C_{CO_2}, \dots, C_{TSP} = carbon mass in CO_2, \dots , and TSP, respectively. It is assumed, in Eq. (1), that the carbon mass in other organic emissions than TNMHC and CH_4 is negligible. A small amount of lighter fluid was used to start burning of some of the solid fuels. In this case, the carbon mass of the lighter fluid was added to the left side of the Eq. (1). Rearranging Eq. (1) leads to:

$$\frac{C_f - C_a}{C_{CO_2}} = 1 + \frac{C_{CO}}{C_{CO_2}} + \frac{C_{CH_4}}{C_{CO_2}} + \frac{C_{TNMHC}}{C_{CO_2}} + \frac{C_{TSP}}{C_{CO_2}}$$

$$= 1 + K, \quad (2)$$

where

$$K = \frac{C_{CO}}{C_{CO_2}} + \frac{C_{CH_4}}{C_{CO_2}} + \frac{C_{TNMHC}}{C_{CO_2}} + \frac{C_{TSP}}{C_{CO_2}}, \quad (3)$$

where C_{CO}/C_{CO_2} , C_{CH_4}/C_{CO_2} , C_{TNMHC}/C_{CO_2} , C_{TSP}/C_{CO_2} are called the emission ratios of CO, CH_4 , TNMHC, and TSP-C, respectively. K is the sum of the emission ratios (relative to CO_2) for CO, CH_4 , etc. and is assumed to represent all products of incomplete combustion (PICs). Thus,

$$C_{CO_2} = \frac{C_f - C_a}{1 + K}. \quad (4)$$

Since the emission factor, E_m , for a pollutant is defined as mass of the pollutant emitted per mass of fuel burned (M), the emission factor for carbon in CO_2 (CO_2 -C) can be calculated from the following equations,

$$E_{m,CO_2-C} = \frac{C_f - C_a}{(1 + K)M} \quad (5)$$

and the emission factor of CO_2 is

$$E_{m,CO_2} = \frac{C_f - C_a}{(1 + K)M} f_{CO_2}, \quad (6)$$

where f_{CO_2} is a conversion factor from carbon mass to the compound mass. Since the molecular weight of CO_2 is 44 and that of carbon is 12, $f_{CO_2} = 3.67$. The emission factors for any other pollutant found in the flue gas, X , can now be calculated easily from the CO_2 emission factor (E_{m,CO_2}) and its molar emission ratio to CO_2 :

$$E_{m,X} = (E_{m,CO_2}) \frac{C_X}{C_{CO_2}} \cdot \frac{(MW)_X}{44} \quad (7)$$

where $E_{m,X}$ is the emission factor of X , C_X is the molar concentration of X , and $(MW)_X$ is the molecular weight of X .

One of the advantages of using the carbon balance model to determine emission factors is that this approach requires the measurement of emission ratios, but not absolute carbon mass, of the emitted airborne species. This means that the sampling probe position can be relatively flexible in the flue gas stream, because it can be reasonably assumed that all airborne pollutants experience the same dilution factor at a sampling position in the flue gas.

Since different amounts of fuels are needed for the same cooking task using different fuel/stove combinations, task-based emission factors (mass of pollutant per cooking task) rather than the fuel mass based is a better performance index to compare the air pollution potential of different fuel/stove combinations (Joshi et al., 1989; Zhang and Smith, 1996). In two previous papers, the standard cooking task was defined as one that would deliver 879 kJ heat to the cooking vessel (Ahuja et al., 1987; Joshi et al., 1989). However, "standard task" is

a rather slippery concept, there is obviously no “gold” definition for it. We think emission factor per “delivered energy” (rather than per cooking task, as used conventionally) is more appropriate to use for the comparison of emissions among different stoves. In this paper, therefore, we report the task-based emission factors as mass of a pollutant emitted per unit energy (1 MJ) delivered to the pot. We chose 1 MJ as unit energy because it is still true that 1 MJ of delivered heat is roughly what a typical one-pot family meal would require in traditional biomass-using household (Ahuja et al., 1987). The conversion from emissions per kg of fuel to emissions per 1 MJ delivered energy can be achieved using the following equation:

$$E_e = \frac{E_m}{H \cdot \eta}, \quad (8)$$

where E_e is the delivered-energy-based emission factor (mass per 1 MJ delivered energy); H is the fuel energy content or calorific value (MJ/kg), and η is the stove thermal efficiency (%). H and η were measured in the study and reported in Table 5.

2.3. Experimental design and sampling approach

Cooking is not a steady-state process. The most common cooking practices have different phases for fuel burning rate (power), namely high and low power phase. Unlike for gaseous fuels, the emission characteristics for solid fuels vary at different times during the burn (Smith et al., 1993; Cooper and Malek, 1982). Hence, it is necessary to choose a burn cycle that is reasonably close to the common cooking practice in the field. For the present study the “water boiling test”, a procedure developed as a standard international method to compare the efficiencies of different stoves, was used with slight modification (VITA, 1985). The water boiling test is a relatively short, simple simulation of common cooking procedure in which a standard quantity of water is used to simulate food. The test includes “high power” and “low power” phases. The high power phase involves heating the standard quantity of water from the ambient temperature to boiling temperature as rapidly as possible. The low power phase follows in which the power is reduced to the lowest level needed to keep the water simmering. Except for coal burning, the burn cycles ranged from 35 to 60 min for all other types of fuel/stove combinations. Coal burning needs a longer cycle due to the slow burn rate especially at the initial phase of coal burning. A pot containing known amount of water was placed on a tested stove during the entire burn cycle. Collected data included those (e.g., water temperature changes, amount of vapor generated, amount of fuel burned) necessary to determine thermal parameters such as burn rate and over-all thermal efficiency of each fuel/stove

combination. This procedure, therefore, has the added advantage of enabling simultaneous measurement of emissions and thermal parameters in a systematic and standard manner.

Three successful tests with complete burn cycles were conducted for each fuel/stove combination. Airborne products of combustion were collected using a sampling configuration which included, from up-stream to downstream of the sampling train, a stainless-steel probe, a filter holder, a pump (SKC Inc., USA), and a clean Tedlar bag (SKC Inc., USA). For stoves with flue, the probe was inserted in the flue. Using this sampling method may cause a measurement error because not all the emissions were passing through the flue during a complete burn cycle. A better experimental design was conducted in India by placing the flue under a hood inside which the sample was taken. However, we did not observe a significant difference between the results obtained using the two different sampling arrangements to test a coal stove. For those having no flues, the stoves were placed under a hood built for the test purpose and the probe was placed inside a hood exhaust duct. The hood method has been used previously in studies of unvented cookstoves and kerosene space heaters (Davidson et al., 1986; Lionel et al., 1986; Ballard-Tremeer and Jawurek, 1996). Filters employed to collect TSP were heat-treated 37-mm quartz fiber filters (Pallflex Products Co., USA). The flow rate of sampling pump was adjusted to fill one or two 80-liter Tedlar bags throughout a whole burn cycle (i.e., from fire start to fire extinction). Indoor background samples were collected at stove mouth height near the door using the same sampling configuration.

An aliquot of air samples (1 ml) was taken using a gas-tight syringe out of a filled 80-l Tedlar bag for immediate analyses of CO₂, CO, CH₄, and TNMHC in an analytical lab located in a building next to the simulated kitchen. When immediate analyses were not possible, about 2 l of air was transferred from the 80-l Tedlar bag to a small (4-l capacity) Tedlar bag using a SKC pump and Teflon tubing. All small bags were analyzed within 3 days of sample collection. Based on our test results, no significant losses of the analyzed compounds occurred from a 3-day storage in small Tedlar bags. Another aliquot of samples was taken using a pump at a flow rate of 0.2–0.4 l min⁻¹ and Teflon tubing out of the 80-l Tedlar bag for SO₂ and NO_x analysis, respectively. SO₂ was collected using an impinger containing a solution of formaldehyde, potassium biphthalate, and Na₂-CDTA buffer solution. NO_x was collected using a CrO₃ tube and an impinger containing a solution of *p*-aminobenzenesulfonic acid, acetic acid, and naphthyl-ethylene-diamine-hydrochloride (NEDA) (Saltzman reagent). The CrO₃ tube, placed in the up-stream of the impinger, was to oxidize NO to NO₂. These were standard methods for sampling ambient SO₂ and NO_x (SEPA, 1992).

2.4. Sample analysis methods

Time, temperature, and the weight of water, fuel, and char were recorded at the beginning and end of the high and low cooking phases. For gaseous fuels, the volume of gas consumed was recorded during each experiment. Fuel energy contents and moisture contents were analyzed by a governmental laboratory at the Center for Fuel Quality Tests and Control in Beijing. Fuel, ash, and char samples were also analyzed for carbon, sulfur, ash and nitrogen contents by the same laboratory using standard fuel analysis methods. TSP concentrations were determined gravimetrically using the particle mass collected on the filters and the sampling volumes. One filter for each fuel/stove combination was analyzed for carbon content of TSP using a thermal-optical carbon analysis technique at Sunset Laboratory, Oregon, USA (Johnson et al., 1981; Turpin et al., 1990).

A system of gas chromatograph – flame ionization detector (FID) – methanizer was employed for analysis of CO₂, CO, and CH₄. In this system, a carbonsphere-packed column was used to separate these three compounds. The separated CO and CO₂ were converted by hydrogen (H₂) at 375°C in a nickel catalytic device (the methanizer) to CH₄ which was then determined by the FID. TNMHC was measured by subtracting CH₄ from the total hydrocarbon (THC) which was determined as CH₄ concentration using a FID and a blank GC column. The small air peak was corrected from THC peak during the analysis.

Approximately 30 min after SO₂ collection, the solution in the impinger was transferred to a volumetric flask and treated (by sequentially adding) with sulfuric acid, sodium hydroxide, and para-rosaniline. After sitting at 20 ± 2°C for 15 min, the treated solution was analyzed colorimetrically at a wave length of 575 nm. Fresh solutions of Na₂SO₃, with known SO₂ equivalent concentrations, were used for constructing the calibration curves (SEPA, 1992).

After sampling, the solution in each NO_x impinger was analyzed colorimetrically at a wave length of 540 nm. High purity sodium nitrite (NaNO₂: > 99.9%) was used to make standard solutions and calibration curves. Gaseous NO_x concentrations were calculated based upon a known conversion factor from gaseous NO₂ concentration to aqueous NO₂⁻ concentration (0.76 for the absorbing solution used in this study) (SEPA, 1992).

2.5. Quality controls

For each fuel/stove combination, preliminary experiments were conducted to standardize the burn cycle and minimize the natural variability due to differences in operator behavior. These trial runs were repeatedly conducted, prior to the three planned tests for each fuel/stove combination, until a satisfactory method

precision (roughly < 20% RSD for major parameters measured) was obtained. Solid fuels were collected in one lot, sun-dried and covered with plastic sheet and kept out of sunlight to avoid any changes in moisture. After each experiment, the doors and windows of the simulated kitchen were opened. Several exhaust fans and side fans were switched on to clean the room properly. Char and ash remaining in each experiment for solid fuels were covered with aluminum foil and labeled for subsequent weight and carbon analysis. All Tedlar bags and Teflon tubing used in each experiment were flushed adequately with compressed clean air for at least three times before each use. Tedlar bags and Teflon tubing used for low-grade fuels such as biomass fuels and coals were not used again. After each fuel/stove combination was tested, the probe was cleaned with DI water and ethanol.

The quartz fiber filters used were baked at 600°C in a clean oven for 2 h to remove any carbon impurities. Each filter was placed in a clean petri dish during transport and storage. Pre-sampling and post-sampling weighing of filters were made using an electronic microbalance with a sensitivity of 10⁻⁵ g. Prior to weighing, all filters were equilibrated in a weighing room for at least 24 h with constant humidity and temperature. Approximately, one blank filter was used for every batch of 10 samples. The blank filters were heat-treated and weighed in the same fashion. The difference between any two times of blank filter weighing was within ± 10%. The exposed filters, placed individually in a petri dish, had been stored in a freezer before they were sent for carbon analyses.

For GC analysis, two or more injections were made for each sample to insure a RSD < 10%. Calibration curves for all measured compounds were made daily and had linear regression R² > 0.99. For every batch of 10 to 15 samples, a standard was spiked to assure that the peak area reading was within ± 10% of the previous reading. The detection limit was about 0.7–1.0 ppm for CO₂, CO, CH₄, and THC. The SO₂ and NO_x analyses followed the similar QC procedure: Calibration curves were made freshly on a daily basis. All calibration curves had R² > 0.99. For every batch of 10 samples, a reference solution (blank) and a standard solution were spiked to assure a ± 10% reproducibility. The method detection limit was 20 µg/m³ for SO₂ and 1.6 µg/m³ for NO_x.

At least one set of parallel sampling of flue gas was conducted for each fuel type. Results from these replicate samples were generally < 20% RSD for the measured species in flue gases.

3. Results and discussion

3.1. Emission ratios

Molar emission ratios to CO₂ are shown in Table 3 for the 28 fuel/stove combinations. The ratios were

Table 3
Molar emission ratios to CO₂-sorted by K

Fuel/stove ^a	K		CO		CH ₄		C _{TNMHC} ^b		C _{TSP} ^b		NO _x ^b		SO ₂ ^b	
	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)
CoalGas-trad	7.48E-05	31	nd	na	nd	na	1.15E-06	173	7.36E-05	31	9.22E-04	11	6.37E-04	43
NaturalGas-trad	2.38E-04	101	1.20E-04	173	nd	na	9.56E-05	100	2.25E-05	76	8.03E-04	28	2.97E-07	118
NaturalGas-IR	2.49E-04	86	nd	na	3.12E-05	173	1.78E-04	119	4.02E-05	26	1.60E-04	45	nd	na
Kero-press	4.31E-03	32	3.78E-03	48	8.74E-06	128	4.87E-04	79	3.80E-05	50	4.72E-04	8	2.44E-06	87
Kero-wick	4.78E-03	21	4.38E-03	23	3.84E-05	60	3.46E-04	34	1.69E-05	33	1.89E-04	56	7.29E-06	87
LPG-trad	5.68E-03	74	1.18E-03	142	4.83E-04	173	3.90E-03	95	1.16E-04	172	1.01E-03	7	nd	na
LPG-IR	1.49E-02	73	9.64E-03	60	2.55E-04	74	4.98E-03	99	2.38E-06	40	2.49E-05	11	6.61E-08	173
CoalBriq-metal	1.99E-02	71	1.96E-02	70	3.72E-05	88	2.10E-04	169	1.21E-05	18	5.98E-04	54	5.28E-05	70
CoalBriq-metal-v	2.33E-02	53	2.32E-02	53	2.83E-05	123	1.31E-05	131	7.47E-05	15	1.63E-04	39	7.30E-04	55
Wood-Brick-v	2.81E-02	13	2.38E-02	19	1.05E-03	28	3.38E-04	85	2.91E-03	15	3.23E-04	94	nd	na
Honey-imp	3.45E-02	61	3.44E-02	61	nd	na	7.30E-06	91	5.67E-05	119	1.52E-04	51	2.14E-05	169
Honey-metal-v	3.65E-02	21	3.65E-02	21	1.09E-05	14	4.45E-07	173	2.66E-05	48	1.70E-04	40	5.20E-05	58
Honey-metal	4.20E-02	41	4.19E-02	41	2.04E-05	173	3.29E-05	136	3.27E-05	17	1.63E-04	11	2.99E-05	90
Honey(s)-metal-v	5.11E-02	44	4.51E-02	38	4.04E-03	96	1.48E-03	149	4.80E-04	118	4.38E-04	35	2.30E-03	5
Coal-metal-v	6.73E-02	53	5.64E-02	42	3.82E-03	121	1.86E-03	111	5.21E-03	117	1.61E-04	66	2.10E-04	149
Wood-India	6.89E-02	37	5.18E-02	31	4.52E-03	47	5.20E-03	81	7.39E-03	50	5.53E-04	55	1.33E-06	113
Maize-Brick-v	6.97E-02	56	5.51E-02	51	3.84E-03	56	9.39E-03	86	1.35E-03	49	1.05E-03	33	8.80E-06	63
Coal-metal	7.03E-02	na	4.43E-02	na	1.13E-02	na	3.53E-03	na	1.12E-02	na	5.79E-05	na	4.07E-05	na
Coal(s)-metal-v	7.37E-02	1	6.59E-02	2	7.44E-03	11	2.95E-04	13	1.08E-04	45	1.79E-03	1	6.81E-03	1
Brush-Brick-v	8.12E-02	29	7.02E-02	26	3.84E-03	52	1.97E-03	92	5.19E-03	18	1.25E-03	23	2.36E-06	137
Wheat-Brick-v	9.53E-02	38	7.36E-02	33	5.54E-03	46	9.91E-03	59	6.24E-03	59	8.13E-04	71	1.59E-05	50
WashCoal-metal-v	9.64E-02	18	5.91E-02	15	1.67E-02	35	3.02E-03	76	1.76E-02	28	6.57E-05	112	2.76E-04	146
Wood-Imp-v	9.78E-02	58	6.96E-02	54	5.93E-03	70	1.25E-02	89	9.79E-03	43	3.37E-04	68	1.39E-05	98
Coal-brick-v	1.24E-01	34	1.21E-01	34	1.03E-03	88	8.70E-04	91	1.42E-03	25	1.03E-03	43	5.50E-05	84
Brush-India	1.36E-01	14	1.05E-01	16	9.82E-03	19	1.14E-02	18	9.72E-03	13	1.06E-03	7	1.15E-06	173
Brush-imp-v	1.39E-01	na	1.10E-01	na	1.45E-02	na	6.15E-03	na	8.59E-03	na	9.84E-04	na	0.00E+00	na
Maize-imp-v	1.64E-01	43	1.32E-01	39	1.62E-02	62	9.02E-03	74	6.70E-03	40	5.52E-04	45	1.29E-04	89
Wheat-imp-v	4.14E-01	46	3.02E-01	31	2.87E-02	89	4.08E-02	101	4.26E-02	69	2.30E-04	64	4.83E-05	173

^aSee Table 1 for fuel/stove codes.

^bBased on 25°C, 1 atm, 1 mg m⁻³ = (m.w./24.5)(ppm) where m.w. = 12 for C_{TNMHC} and C_{TSP}, m.w. = 46 (NO₂) for NO_x, and m.w. = 64 for SO₂.

Key: nd = not detected or background level ≥ flue gas concentration; na = only one measurement made or not applicable; K = sum of molar ratios for CO, CH₄, TNMHC and TSP-C. c.v. = (standard deviation)/mean, derived from three tests for each fuel/stove combination.

calculated using net concentrations of each reported compound and net concentrations of CO_2 (net concentration equals flue gas concentration minus background concentration). Reported in Table 3 are the means and coefficients of variation (c.v.) derived from three successful tests for each fuel/stove combination. Because C_{TSP} , SO_2 and NO_x were measured as mg/m^3 , the molar ratios were calculated using conversion factors based on 25°C and 1 atmosphere condition (see footnotes of Table 3). The concentrations of C_{TSP} were obtained using measured carbon content of TSP (%) and TSP concentration (mg/m^3). K is the sum of emission ratios of all other carbon-containing species than CO_2 in the flue gas and represents the emission ratio for total product of incomplete combustion (PIC). Since the ideal combustion would convert all carbon in the fuel burnt to CO_2 solely, K is inversely proportional to the nominal combustion efficiency (Smith et al., 1999). The results shown in Table 3 indicate that the coal gas and natural gas had highest combustion efficiencies, followed by kerosene and LPG, then followed by various coals and fuel wood, and then by crop residues and brush wood, with a very wide range for K (nearly 4 order of magnitude).

3.2. Emission factors

The emission factors are reported on a fuel mass basis in Table 4 and on a delivered energy basis in Table 5. (The thermal efficiency value, η , of each tested fuel/stove combination and the energy content, H , of each tested fuel are reported in Table 5 as well.) The means and c.v. values were derived from the three successful experiments for each of the tested fuel/stove combinations. The PIC emission factors are the summation of the emission factors for CO , CH_4 , C_{TNMHC} , and TSP. (SO_2 and NO_x are not included because they do not contain carbon.) For each of the 6 wood/stove combinations tested, two values of emission factors are reported in Tables 4 and 5: the smaller value being called instant emission factor and the larger value (reported in parentheses) being called ultimate emission factor. The reasoning for reporting the two values is as follows.

During combustion of wood in a cookstove, fuel carbon is mainly converted to TSP and gases including CO_2 , CO , and volatile organic compounds. A small fraction of fuel carbon is diverted into bottom ash or remains as the partially burned material, char. In determining instant emission factors for the airborne components, we subtract char carbon from the fuel carbon in the carbon balance equation. The emission factors determined in this way, thus, represent the emission measures for instant wood combustion, assuming the char generated during the combustion will not be burnt later.

In reality, users of wood cookstoves usually save unburned char to be burned along with wood at the next

meal or extracted and stored for later use to cook a meal entirely with the char or to use in the winter in small personal braziers for heating. In this situation, the char carbon generated through primary wood combustion would be ultimately converted to airborne carbon species entering the atmosphere. The amount of each airborne species associated with the secondary char combustion should be added to the instant emission factors to truly represent the ultimate emission measures. Since we realized this issue after having finished all the field experiments in which no tests for char combustion had been planned, we decide to use the data from several low-quality charcoal combustion measurements for calculating the ultimate emission factors, assuming that the char would have the same emission factors as the low-quality charcoal. The tests for charcoal/stove combinations were conducted in India (Smith et al., 1999). The results show that every gram of char (or 0.81 g carbon in the char) generated through the primary wood combustion would be converted during secondary char burning to: 0.66 g C_{CO_2} (or 2.42 g CO_2); 0.12 g C_{CO} (or 0.28 g CO); 0.0059 g C_{CH_4} (or 0.0079 g CH_4); 0.0068 g C_{TNMHC} ; 0.0020 g C_{TSP} (or 0.0024 g TSP); and 0.0099 g carbon in remaining char and ash. Note that the remaining char produced during the secondary combustion had low carbon contents (< 21% by weight) and small quantity, i.e., too poor to be attractive as fuel. It seem justifiable, therefore, to consider this as the solid carbon that becomes part of the disposed ash and char and is thus sequestered from the atmosphere, if not permanently, at least for long periods.

The results indicate that the difference between instant emission factor and ultimate emission factor was much smaller for CO_2 than for PIC, suggesting that flaming combustion of wood would be more complete than charcoal burning at a relatively steady rate.

For solid fuels, emission factors are sometimes reported on a dry fuel mass basis. Since the fuel moisture content for each tested solid fuel (crop residue, wood, and coal) was measured (see Table 2), the fuel-mass-based emission factors shown in Table 4 can be easily converted to dry-fuel-mass-based emission factors.

3.3. Implications and potential applications of the database

Use of this database can be expected to reduce uncertainties in estimating GHG emissions from combustion of domestic fuels in China and in other countries with similar situations, especially when stove-specific fuel consumption data are available. Unfortunately, regional or national fuel data have often not been stove-specific, perhaps partly due to the lack of such a detailed database of emission factors. We thus hope that these fuel/stove specific emission factors will stimulate the collection of stove-specific fuel data in the future to further improve the emission estimates.

Table 4
Emission factors (gram of compound per kg of fuel) — sorted by PIC

Fuel/stove ^a	PIC		CO ₂		CO		CH ₄		CTNMHC ^b		TSP		NO _x		SO ₂	
	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)	Mean	c.v.(%)
Coal/Gas-trad	2.00E-01	31	1.85E + 03	< 1	nd	na	nd	na	5.79E-04	173	1.99E-01	31	1.78E + 00	11	1.71E + 00	43
Natural/Gas-IR	4.09E-01	30	3.44E + 03	< 1	nd	na	3.91E-02	173	1.67E-01	119	2.03E-01	26	5.77E-01	45	nd	na
Natural/Gas-trad	4.66E-01	115	3.44E + 03	< 1	2.63E-01	173	nd	na	8.97E-02	100	1.13E-01	76	2.89E + 00	28	1.49E-03	118
LPG-trad	6.65E-00	74	3.09E + 03	< 1	2.31E + 00	142	5.40E-01	173	3.28E + 00	95	5.24E-01	172	3.26E + 00	7	nd	na
Kero-press	8.11E-00	39	3.13E + 03	< 1	7.51E + 00	47	9.94E-03	128	4.15E-01	79	1.74E-01	50	1.54E + 00	8	1.11E-02	87
Kero-wick	9.11E-00	22	3.12E + 03	< 1	8.70E-00	23	4.36E-02	60	2.95E-01	34	7.74E-02	33	6.18E-01	56	3.31E-02	87
CoalBriq-metal-v	1.93E + 01	32	1.39E + 03	20	1.91E + 01	32	1.20E-02	109	5.43E-03	130	1.71E-01	18	2.48E-01	53	1.59E + 00	64
CoalBriq-metal	2.00E + 01	73	1.60E + 03	13	1.99E + 01	72	2.05E-02	90	9.28E-02	169	3.22E-02	25	9.82E-01	55	1.20E-01	67
LPG-IR	2.31E + 01	66	3.06E + 03	1	1.87E + 01	59	2.82E-01	73	4.12E + 00	98	1.06E-02	40	7.95E-02	10	2.97E-04	173
Wood-Brick-v ^c	2.59E + 01	15	1.56E + 03	3	2.36E + 01	18	5.96E-01	29	1.41E-01	84	1.54E + 00	13	5.24E-01	92	nd	na
	(3.72E + 01)	24	(1.62E + 03)	1	(3.08E + 01)	21	(2.45E + 00)	55	(1.74E + 00)	75	(2.18E + 00)	28	(5.44E-01)	94	nd	na
Maize-Brick-v	4.65E + 01	51	1.16E + 03	3	4.03E + 01	48	1.60E + 00	54	2.93E + 00	84	1.68E-00	46	1.27E-00	34	1.50E-02	64
Wood-India ^c	4.98E + 01	16	1.38E + 03	24	4.33E + 01	15	2.09E + 00	17	1.70E + 00	53	2.67E + 00	45	8.53E-01	69	2.22E-03	97
	(8.88E + 01)	73	(1.55E + 03)	4	(6.95E + 01)	123	(7.92E + 00)	123	(6.84E + 00)	134	(4.53E + 00)	74	(9.05E-01)	58	(2.91E-03)	110
Honey-metal-v	5.91E + 01	20	2.55E + 03	5	5.89E + 01	20	1.01E-02	18	3.24E-04	173	2.17E-01	48	4.47E-01	37	1.90E-01	57
Honey-imp	5.94E + 01	59	2.71E + 03	2	5.89E + 01	59	nd	na	5.44E-03	92	4.84E-01	119	4.34E-01	52	8.28E-02	169
Honey-metal	6.65E + 01	35	2.55E + 03	9	6.62E + 01	35	1.77E-02	173	2.48E-02	140	2.71E-01	27	4.32E-01	3	1.15E-01	93
Wheat-Brick-v	7.20E + 01	31	1.32E + 03	5	6.11E + 01	27	2.62E + 00	41	3.50E + 00	54	4.73E + 00	54	1.14E + 00	72	3.08E-02	52
Brush-Brick-v ^c	7.21E + 01	25	1.50E + 03	3	6.66E + 01	24	2.07E + 00	49	0.79E-01	89	2.65E + 00	15	1.95E + 00	23	5.24E-03	137
	(7.45E + 01)	26	(1.62E + 03)	2	(6.82E + 01)	25	(2.42E + 00)	51	(1.09E + 00)	81	(2.78E + 00)	17	(1.96E + 00)	23	(5.26E-03)	137
Honey(s)-metal-v	9.20E + 01	41	2.38E + 03	3	6.84E + 01	38	3.46E-00	97	9.60E-01	149	6.22E-01	118	1.09E + 00	35	7.99E + 00	7
Wood-Imp-v ^c	7.75E + 01	51	1.50E + 03	6	6.53E + 01	49	3.16E + 00	67	4.95E + 00	86	4.10E + 00	37	5.43E-01	71	3.08E-02	96
	(8.21E + 01)	50	(1.52E + 03)	5	(6.84E + 01)	49	(3.83E + 00)	66	(5.56E + 00)	79	(4.32E + 00)	36	(5.49E-01)	71	(3.14E-02)	97
Coal(s)-metal-v	9.21E + 01	na	2.51E + 03	na	7.07E + 01	na	1.03E + 01	na	2.41E-00	na	8.73E + 00	na	1.52E-01	na	1.48E-01	na
Coal-metal	1.00E + 02	37	1.06E + 03	7	8.77E + 01	34	6.09E + 00	59	2.52E-00	71	4.02E + 00	35	2.06E-01	52	2.06E-01	92
Maize-Imp-v	1.04E + 02	43	2.70E + 03	9	9.55E + 01	38	3.52E + 00	114	1.30E-00	104	4.13E + 00	111	4.70E-01	68	8.76E-01	152
Coal-metal-v	1.09E + 02	14	1.42E + 03	2	9.5E + 01	15	5.05E + 00	18	4.41E + 00	16	4.15E + 00	14	1.57E + 00	8	2.33E-03	173
Brush-India ^c	(1.13E + 02)	12	(1.43E + 03)	2	(9.8E + 01)	14	(5.65E + 00)	20	(4.94E + 00)	21	(4.36E + 00)	14	(1.59E + 00)	7	(2.34E-03)	173
Brush-Imp-v ^c	1.12E + 02	na	1.41E + 03	na	9.81E + 01	na	7.40E + 00	na	2.36E + 00	na	4.60E + 00	na	1.45E + 00	na	nd	na
	(1.19E + 02)	na	(1.43E + 03)	na	(1.03E + 02)	na	(8.32E + 00)	na	(3.09E + 00)	na	(4.97E + 00)	na	(3.09E + 00)	na	nd	na
WashCoal-metal-v	1.20E + 02	15	2.38E + 03	6	8.92E + 01	13	1.43E + 01	32	1.99E-00	80	1.42E + 01	26	1.67E-01	114	9.80E-01	147
Coal-brick-v	1.69E + 02	39	2.13E + 03	15	1.66E + 02	39	8.60E-01	96	5.19E-01	87	1.60E + 00	40	2.21E + 00	35	1.84E-01	97
Wheat-Imp-v	2.15E + 02	30	9.59E + 02	18	1.79E + 02	22	8.97E + 00	90	9.62E-00	102	1.80E + 01	56	2.29E-01	59	8.16E-02	173

^aSee Table 1 for fuel/stove codes.

^bGram of carbon in TNMHC.

^cIncluding instant emission factor and ultimate emission factor in ().

Key: nd = not detected or background level \geq flue gas concentration; na = only one measurement made or not applicable; PIC = CO + CH₄ + TNMHC-C + TSP.
c.v. = (standard deviation)/mean, derived from three tests for each fuel/stove combination.

Table 5
Emission factors (gram of compound per 1 MJ delivered energy) — sorted by PIC

Fuel/stove ^a	Stove efficiency		Fuel H	PIC	CO ₂		CO		CH ₄		C ₁₀ H ₁₆ ^b		TSP		NO _x		SO ₂		
	Mean (%)	c.v. (%)			Mean	c.v. (%)	Mean	c.v. (%)	Mean	c.v. (%)	Mean	c.v. (%)	Mean	c.v. (%)	Mean	c.v. (%)	Mean	c.v. (%)	Mean
CoalGas-trad	45.89	10	43.756	9.82E-03	22	9.25E+01	10	nd	na	nd	na	2.96E-05	173	9.79E-03	22	8.86E-02	3	8.34E-02	33
NaturalGas-IR	60.92	<1	51.297	1.31E-02	60	1.10E+02	<1	nd	na	1.24E-03	173	5.35E-03	119	6.49E-03	27	1.85E-02	45	nd	na
NaturalGas-trad	53.69	2	51.297	1.69E-02	114	1.25E+02	2	9.51E-03	173	nd	na	3.29E-03	101	2.09E-03	76	1.05E-01	29	5.37E-05	117
LPG-trad	45.22	5	49.003	2.98E-01	72	1.40E+02	5	9.96E-02	141	2.31E-02	173	1.51E-01	99	2.49E-02	172	1.48E-01	12	nd	na
Kero-press	45.89	7	43.260	4.08E-01	40	1.58E+02	7	3.78E-01	49	5.20E-04	127	2.10E-02	82	8.68E-03	48	7.77E-02	3	5.83E-04	87
Kero-wick	44.76	7	43.260	4.68E-01	15	1.62E+02	8	4.46E-01	17	2.20E-02	58	3.20E-03	33	4.05E-03	39	3.24E-02	59	1.74E-03	87
LPG-IR	42.13	21	49.003	1.28E+00	90	1.53E+02	23	1.03E+00	84	1.58E-02	96	2.40E-01	118	5.40E-04	45	4.05E-03	35	1.29E-05	173
CoalBriq-metal	37.11	3	13.852	3.92E+00	74	3.12E+02	15	3.89E+00	74	3.97E-03	91	1.83E-02	169	6.27E-03	26	1.91E-01	56	2.33E-02	67
CoalBriq-metal-v	27.23	35	13.852	5.46E+00	42	4.15E+02	48	5.40E+00	48	2.94E-03	84	2.05E-03	141	4.91E-02	35	7.54E-02	61	5.12E-01	78
Honey-imp	46.61	4	19.210	6.58E+00	58	3.03E+02	6	6.52E+00	58	nd	na	6.22E-04	94	5.28E-02	116	4.89E-02	55	9.16E-03	169
Honey(s)-metal-v	44.06	8	20.325	8.27E+00	45	2.67E+02	5	7.68E+00	41	4.05E-01	97	1.12E-01	149	7.20E-02	120	1.21E-01	34	8.93E-01	6
Wood-Brick-v ^c	13.13	19	16.246	1.22E+01	12	7.50E+02	12	7.50E+02	10	2.94E-01	43	7.54E-02	101	7.59E-01	36	2.19E-01	91	nd	na
Honey-metal	23.41	11	19.210	1.47E+01	31	5.73E+02	18	1.46E+01	32	4.13E-03	173	5.85E-03	146	6.16E-02	35	9.68E-02	11	2.59E-02	99
Wood-India ^c	21.42	37	16.246	1.68E+01	59	4.58E+02	59	1.46E+01	60	6.63E-01	38	5.31E-01	50	9.71E-01	84	2.66E-01	64	5.40E-04	101
Honey-metal-v	16.16	6	19.210	1.91E+01	21	8.24E+02	11	1.90E+01	21	3.29E-03	23	1.11E-04	173	7.02E-02	48	1.42E-01	31	6.02E-02	55
Wood-imp-v ^c	23.96	9	16.246	2.05E+01	60	3.88E+02	8	1.72E+01	58	8.31E-01	73	1.33E+00	94	1.08E+00	47	1.36E-01	71	7.49E-03	92
Coal-metal	14.27	na	27.301	2.36E+01	na	6.43E+02	na	1.81E+01	na	2.65E+00	na	6.19E-01	na	2.24E+00	na	3.89E-02	na	3.81E-02	na
Coal-metal-v	17.64	75	27.301	2.83E+01	74	7.42E+02	49	2.56E+01	66	1.08E+00	150	3.78E-01	144	1.25E+00	147	1.19E-01	97	1.16E-01	114
Maize-Brick-v	11.00	17	16.083	2.85E+01	69	6.66E+02	16	2.46E+01	67	9.83E-01	71	1.86E+00	99	1.02E+00	66	7.03E-01	21	7.96E-03	57
Maize-imp-v	19.09	8	16.083	3.31E+01	42	3.48E+02	9	2.89E+01	39	2.02E-00	65	8.46E-01	77	1.34E+00	43	2.02E-01	50	6.49E-02	93
Brush-Brick-v ^c	13.74	10	15.333	3.49E+01	35	7.14E+02	8	3.22E+01	34	1.01E+00	59	3.99E-01	98	1.28E+00	24	9.25E-01	21	2.42E-03	139
Brush-India ^c	16.95	2	15.329	4.19E+01	15	5.45E+02	1	3.66E+01	16	1.95E+00	19	1.70E+00	17	1.60E+00	13	6.06E-01	7	9.11E-04	173
WashCoal-metal-v	9.31	8	30.054	4.33E+01	14	5.52E+02	1	3.78E+01	15	2.18E+00	21	1.90E+00	22	1.67E+00	13	6.13E-01	6	(9.17E-04)	149
Coal-brick-v	17.02	45	27.301	4.49E+01	64	5.42E+02	55	4.41E+01	63	2.67E-01	114	1.45E-01	83	4.30E-01	78	5.03E-01	35	5.59E-02	123
Brush-imp-v ^c	14.41	na	15.329	5.09E+01	na	6.36E+02	na	4.44E+01	na	3.35E+00	na	1.07E+00	na	2.08E+00	na	6.55E-01	na	nd	na
Wheat-Brick-v	9.69	11	14.023	5.47E+01	43	9.76E+02	7	4.63E+01	40	2.01E+00	52	2.70E-00	66	3.65E+00	66	8.10E-01	70	2.23E-02	48
Coal(s)-metal-v	6.82	<1	20.939	6.45E+01	1	1.44E+03	<1	6.04E+01	2	3.89E+00	11	1.16E-01	13	9.42E-02	45	2.69E+00	1	1.43E+01	1
Wheat-imp-v	15.40	8	14.023	1.01E+02	36	4.47E+02	22	8.39E+01	29	4.19E+00	94	4.56E+00	108	8.48E+00	62	1.10E-01	64	3.88E-02	173

^aSee Table 1 for fuel/stove codes.

^bGram of carbon in TNMHC.

^cIncluding instant emission factor and ultimate emission factor in ().

Key: nd = not detected or background level > flue gas concentration; na = only one measurement made or not applicable; PIC = CO + CH₄ + TNMHC-C + TSP. c.v. = (standard deviation)/mean, derived from three tests for each fuel/stove combination.

Table 6
Mean, minimum and maximum emission factors of each fuel type (gram of compound per kg of fuel)

Fuel Type		CO ₂	CO	CH ₄	C _{TNMHC} ^a	TSP	NO _x	SO ₂
Wood (Instant Emission)	Min	9.87E + 02	1.97E + 01	3.80E-01	2.57E-02	1.17E + 00	1.23E-01	nd
	Mean	1.45E + 03	5.87E + 01	2.70E + 00	2.27E + 00	3.05E + 00	1.16E + 00	7.91E-03
	Max	1.62E + 03	1.11E + 02	7.40E + 00	9.68E + 00	5.87E + 00	2.77E + 00	6.08E-02
Wood (Ultimate Emission)	Min	1.41E + 03	2.41E + 01	5.37E + 01	2.94E-01	1.51E + 00	1.24E-01	nd
	Mean	1.52E + 03	6.92E + 01	5.06E + 00	4.34E + 00	3.82E + 00	1.19E + 00	8.21E-03
	Max	1.63E + 03	1.23E + 02	1.99E + 01	1.77E + 01	8.73E + 00	2.78E + 00	6.24E-02
Crop Residues	Min	8.34E + 02	2.36E + 01	4.00E-02	4.64E-02	1.12E + 00	3.93E-02	nd
	Mean	1.13E + 03	8.63E + 01	4.56E + 00	4.35E + 00	8.05E + 00	7.00E-01	2.16E-01
	Max	1.37E + 03	2.23E + 02	1.59E + 01	1.97E + 01	2.90E + 01	2.21E + 00	2.64E + 00
Coal	Min	1.07E + 03	1.10E + 01	nd	nd	2.60E-02	8.28E-02	nd
	Mean	2.28E + 03	7.13E + 01	2.92E + 00	6.64E-01	1.30E + 00	9.14E-01	2.67E + 00
	Max	2.91E + 03	2.10E + 02	1.69E + 01	6.90E + 00	1.00E + 01	3.86E + 00	2.05E + 01
Kerosene	Min	3.12E + 03	2.37E + 00	nd	9.18E-02	4.61E-02	2.18E-01	nd
	Mean	3.13E + 03	7.39E + 00	2.48E-02	3.92E-01	1.34E-01	1.10E + 00	2.49E-02
	Max	3.13E + 03	1.09E + 01	6.24E-02	9.21E-01	2.83E-01	1.69E + 00	6.31E-02
Gases	Min	1.85E + 03	nd	nd	nd	nd	7.23E-02	nd
	Mean	2.98E + 03	3.72E + 00	1.37E-01	1.60E-00	2.61E-01	1.76E + 00	3.30E-01
	Max	3.44E + 03	3.13E + 01	1.62E + 00	8.79E + 00	1.62E + 00	4.51E + 00	2.54E + 00

^aGram of carbon in TNMHC.

Key: nd = not detected or background level > flue gas concentration.

When stove-specific fuel data are not available, one may use fuel-specific emission factors for emission estimates. For this purpose, we have summarized in Table 6 the range and mean values of the emission factors measured for the five types of fuels.

The results, verifying the preliminary findings from the pilot study (Smith et al., 1993), show solid fuels (biomass and coal) are typically burned with substantial production of PIC. Some fuel/stove combinations diverted more than 10% (up to 25% for Wheat-Imp-v) of the fuel carbon into PIC. No biomass stove produced less than 4% (ultimate emission) of original fuel carbon as PIC. Greenhouse analyses of human fuel use often assume that renewably harvested biomass fuels do not contribute to global warming, because the released carbon is entirely recycled through photosynthesis in growing biomass that replaces the burned biomass. Even under renewable harvesting, however, the gases released as PIC contribute to global warming because of their higher radiative forcing than CO₂ or their indirect greenhouse effects. (Direct GHGs emitted from fuel combustion include CO₂, CH₄, N₂O and indirect GHGs include CO, NO_x, etc.) (Hayes and Smith, 1994; IPCC, 1990,1995). Thus, biomass fuels have the potential to produce net global warming commitment even when grown renewably due to the contribution of non-CO₂ GHGs.

The results show that all the biomass stoves (as well as most coal stoves) tested had substantially lower thermal efficiencies than those using liquid and gaseous fuels (see

Table 5). In addition, the solid fuels had substantially lower energy contents than liquid and gaseous fuels (see Table 5). As a result, the total PIC and CO₂ emissions per unit delivered energy were substantially greater from burning solid fuels. In general, the ranking follows what has been called the “energy ladder” from lower to higher quality fuels, i.e., emissions decrease and efficiencies increase in the following order: crop residues, brush wood > fuel wood > kerosene > gas (Smith and Liu, 1994). The ranking for coal was largely depending on the specific coal types and stove designs.

The data shown in Table 5 can be used to quantitatively assess both GHG and health benefits that can be brought through switching usage from a lower-quality fuel (stove) to a higher-quality one. For example, using Wood-Brick-v to replace Wheat-Brick-v (i.e., same stove but different fuel type) to perform a same cooking task (i.e., to deliver 1 MJ energy to the pot) would reduce emissions of CO₂ by a factor of 1.3, CH₄ by 1.6, CO by 3.1, TNMHC by 3.1, NO_x by 3.5, TSP by 3.3, and total PIC by 3. Comparing PIC emissions from pairs of traditional and improved biomass stoves using the same type of fuel (wood-Brick-v vs. wood-Imp-v; Brush-Brick-v vs. Brush-Imp-v; Maize-Brick-v vs. Maize-Imp-v; Wheat-Brick-v vs. Wheat-Imp-v), we have found, interestingly, that the improved stoves tend to generate greater PIC emissions (see Table 5). As shown in Table 5, it is true that the improved stoves had higher overall stove efficiencies. However, the overall stove efficiency is the

product of combustion efficiency and heat transfer efficiency. The increase in overall stove efficiency for the so-called improved stoves is often achieved mainly by increasing the heat transfer efficiency (e.g., using tight insulation) which may consequently reduce the combustion efficiency (Smith, 1987). A decreased combustion efficiency could lead to less complete combustion and emit more PICs. Therefore, an integrated assessment of GHG emissions, thermal efficiency, and health impact can provide more balanced, fair, and complete evaluation of stoves and other combustion devices than only considering one of these three important aspects.

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References

- Ahuja, D.R., Joshi, V., Smith, K.R., Venkataraman, C., 1987. Thermal performance and emission characteristics of unvented biomass-burning cookstoves: a proposed standard method for evaluation. *Biomass* 12, 247–270.
- Ballard-Tremere, G., Jawurek, H.H., 1996. Comparison of five rural wood-burning cooking devices: efficiencies and emissions. *Biomass and Bioenergy* 11, 419–430.
- Cofer, W.R., Levine, J.S., Winstead, E.L., Stocks, B.J., Cahoon, D.R., Pinto, J.P., 1993. Trace gas emissions from tropical biomass fires: Yucatan Peninsula, Mexico. *Atmospheric Environment* 27A, 1903–1907.
- Cooper, J.A., Malek, D. (Eds.), 1982. *Residential Solid Fuels: Environmental Impacts and Solutions*. Oregon Graduate Center, Beaverton, Oregon.
- Crutzen, P.J., Andreae, M.O., 1990. Biomass burning in the tropics: impact on atmospheric chemistry and biogeochemical cycles. *Science* 250, 1669–1678.
- Davidson, C., Borrazzo, J.E., Hendrickson, C.T., 1986. Pollutant emission factors for gas stoves: a literature survey. Report CR-812543-01-0. US EPA, Research Triangle Park, NC.
- FAO (Food and Agriculture Organization), 1987. *Yearbook of Forest Products*, Rome, Italy.
- Gu, S., Huang, K., Qiu, D., Smith, K.R., 1991. 100-Million improved cookstoves in China: how was it done? Country Study No. 1, Global Review of Improved Cookstove Programs. Environment and Policy Institute, East–West Center, Honolulu, HI, USA.
- Hayes, P., Smith, K.R., 1994. *The Global Greenhouse Regime: Who Pays?* Earthscan, London, UK.
- IPCC (Inter-governmental Panel on Climate Change), 1997. In: Houghton, J.T. et al., (Ed.), *Guidelines for National Greenhouse Gas Inventories: Reference Manual (Revised 1996)*. Vol. 3. Bracknell, UK.
- IPCC (Inter-governmental Panel on Climate Change), 1995. *Radiative Forcing of Climate Change*, Cambridge University Press, UK.
- IPCC (Inter-governmental Panel on Climate Change), 1990. *Climate Change: the IPCC Scientific Assessment*. Cambridge University Press, UK.
- Jenkins, B.M., Turn, S.Q., Williams, R.B., 1992. Atmospheric emissions from agricultural burning in California: determination of burn fractions, distribution factors, and crop-specific contributions. *Agriculture, Ecosystems and Environment* 38, 313–330.
- Johnson, R.L., Shah, J.J., Cary, R.A., Huntzicker, J.J., 1981. An automated thermal-optical method for the analysis of carbonaceous aerosol in ACS Symposium Series No. 167, *Atmospheric Aerosol: Source/Air Quality Relationships*. Eds., E.S. Macias and P.K. Hopke.
- Joshi, V., Venkataraman, C., Ahuja, D.R., 1989. Emissions from burning biofuels in metal cookstoves. *Environmental Management* 13, 763–772.
- Levine, J.S. (Ed.) 1996. *Biomass Burning and Global Change*. Vol. 1&2, MIT Press, Cambridge, MA, USA.
- Lionel, T., Martin, R.J., Brown, N.J., 1986. A comparative study of combustion in kerosene heaters. *Environmental Science and Technology* 20, 78–85.
- SEPA (State Environmental Protection Administration of China), 1992. *Standardized Environmental Monitoring and Analysis Methods*. SEPA, Beijing, China.
- Smith, K.R., 1987. *Biofuels, Air Pollution, and Health*. Plenum Publishing Corp., Plenum Press, New York.
- Smith, K.R., Liu, Y., 1994. Indoor air pollution in developing countries. In: *Epidemiology of Lung Cancer*, Samet, J.M. (Ed.), Lung Biology in Health and Disease Series. Vol. 74, Marcel Dekker, Inc., New York. p.p. 151–184.
- Smith, K.R., Uma, R., Kishore, V.V.N., Lata, K., Joshi, V., Zhang, J., Rasmussen, R.A., Khalil, M.A.K., Thorneloe, S.A., 1999. *Greenhouse Gases from Small-scale Combustion Devices in Developing Countries, Phase IIa: Household Stoves in India*. Office of Research and Development, US EPA, Washington, DC, USA.
- Smith, K.R., Khalil, M.A.K., Rasmussen, R.A., Thorneloe, S.A., Manegdeg, F., Apte, M., 1993. Greenhouse gases from biomass and fossil fuel stoves in developing countries: a Manila pilot study. *Chemosphere* 26, 479–505.
- Turpin, B.J., Carey, R.A., Huntzicker, J.J., 1990. An in situ, time-resolved analyzer for aerosol and organic carbon. *Journal of Aerosol Science and Technology* 12, 161–167.

- VITA (Volunteers in Technical Assistance, Inc.), 1985. Testing The Efficiency of Wood-burning Cookstoves: International Standards, Arlington, VA, USA.
- Wang, Q.H., 1994. Investigation of rural crop straw utilization in China. *Rural Eco-environment* 10, 67–71.
- Ward, D.E., Hardy, C.C., 1991. Smoke emissions from wildland fires. *Environmental International* 17, 117–134.
- Zhang, J., Smith, K.R., 1996. Hydrocarbon emissions and health risks from cookstoves in developing countries. *Journal of Exposure Analysis and Environmental Epidemiology* 6, 147–161.