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Carbon monoxide from cookstoves in developing countries: 1. Emission factors

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Importance of this Paper. Cookstoves in developing countries are individually small, but so numerous that, depending on emission factors, they could be significant influences on CO inventories. This paper presents a new and comprehensive database of CO emission factors for 56 fuellstove combinations commonly used in developing countries. These include various stoves using animal dung, different species of crop residues and wood, root fuel, charcoal, kerosene, and several types of coals and gases. The chosen fuellstove combinations represent a large fraction of the total in developing countries and a large fraction of biomass stoves used worldwide. Thus the database can be used to improve estimates of national and global CO emission inventories.

Abstract

Cookstoves in developing countries are individually small, but so numerous that, depending on emission factors, they could be significant influences on global and regional carbon monoxide (CO) inventories. This paper presents a new database of CO emission factors for commonly used cookstoves in developing countries. The emission factors were determined using a carbon balance approach for 56 types of fuel/stove combinations in China and India. These include various stoves (e.g., traditional, improved, mud, brick, and metal, with and without chimney) using animal dung, different species of crop residues and wood, root fuel, charcoal, kerosene, and several types of coals and gases. The chosen fuel/stove combinations represent a large fraction of the total in developing countries. Thus, the database can be used to improve estimates of CO emission inventories. The CO emission factors ranged widely, from 3.0×10^{-2} g/kg for the coal gas/traditional stove to 2.8×10^2 g/kg for the charcoal/Angethi stove, nearly 4 orders of magnitude. Since stove efficiencies and fuel energy contents were measured simultaneously, CO emission factors on the basis of a standard

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cooking task (energy delivered) were also determined and reported in this paper. Task-based emission factors are particularly useful for comparing the air pollution potential of different fuel/stove combinations and assessing the impacts of substitutions. © 1999 Elsevier Science Ltd. All rights reserved.

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1. Introduction

It is clear that carbon monoxide (CO) released into the atmosphere plays an important role in global, regional, and urban atmospheric chemistry by affecting the concentration of the hydroxyl radical (OH) and the cycle of tropospheric ozone (O₃) (Seinfeld and Pandis, 1997). CO is considered an indirect greenhouse gas due to its close coupling to atmospheric methane (CH₄), a strong greenhouse gas (IPCC, 1990; Khalil and Rasmussen, 1985). CO is often found to be a toxic air pollutant in urban polluted air and indoor air (Hubbard, 1995; Klouda and Connolly, 1995). Current estimates indicate that combustion of fossil fuels and biomass fuels contributes about 44% (range: 36–78%) of the total global CO budget (Khalil and Rasmussen, 1990). However, the uncertainty of the current estimates is large, partly due to the lack of an accurate database of CO emission factors. In recent years, the wide uncertainty about the emissions of CO and other trace gases 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), agricultural burning, etc. (Jenkins et al., 1992; Crutzen and Andreae, 1990). A significant fraction of all biomass combustion, however, occurs in enclosed or semi-enclosed small-scale devices such as cooking and space-heating devices. For example, about 90% of the firewood and a large fraction of the combusted crop residues are burned in such conditions worldwide, mainly in developing countries (FAO, 1987; Smith et al., 1993). These uses make up 10–15% of global direct human energy use. Such small-scale devices are expected to have different emission factors compared to the open, large-scale combustion. Unfortunately, few measurements have been made to

determine emission factors for these devices in developing countries. Thus, it is necessary to conduct measurements of CO emission factors for a range of fuels and combustion devices to improve estimates of regional, national, and global inventories of CO. This is also needed for estimating the impact on CO emissions and exposures of fuel switching. Estimates of CO exposures resulting from fuel combustion in various cookstoves is presented in Part 2 of our report on CO cookstove emissions, which is also published in this issue.

Experiences have shown that slight changes in combustion conditions in small stoves (e.g., in the fire-tending behavior) can have large impacts on emission factors. Some stoves are so massive that they can not be easily transported. Therefore, it would be more appropriate to conduct tests in the field rather than in a controlled setting such as a chamber. In a typical chamber test, the emission factor of a measured species can be determined by constructing a mass balance model for the measured species only, i.e., one only needs to measure concentrations of the species at the input and output points of the chamber plus the air flow rate. Because air flow rates vary greatly in actual homes, we have applied a carbon balance approach to measure emission factors. This approach does not require the measurement of air flow rate but requires a complete carbon analysis in the fuel, ash and unburned residues, and all airborne emissions.

Carbon balance models have been often used to determine emission factors for large-scale open biomass fires (Delmas et al., 1995; Hurst et al., 1994a, b). In a similar fashion, a carbon balance model has been used to determine emission factors for several cookstoves in a Manila pilot study (Smith et al., 1993). After this carbon balance model was validated in the pilot study, we have conducted the present study in China and India

where total of 56 fuel/stove combinations were tested for emission factors of carbon dioxide (CO₂), CO, CH₄, total non-methane hydrocarbons (TNMHC), some 60 species of hydrocarbons, nitrogen oxides (NO_x), and sulfur dioxide (SO₂). In this paper, we only report the methodology used and a comprehensive database of CO emission ratios and CO emission factors both on a per-fuel-mass basis and on a per-cooking-task basis. All other data required for the carbon balance approach, as well as details in methods and quality assurance, are reported elsewhere (Smith et al., 1999; Zhang et al., 1997; Smith et al., 1995).

2. Methodology

As shown in Table 1 and Table 2, 56 fuel/stove combinations were tested in China and India, 28 in each country. In India, all of the fuel/stove combinations were tested in a simulated village house located in suburban Delhi. In China, the vast majority of the fuel/stove combinations were tested in a simulated village house located in suburban Beijing, while those stoves using piped gas fuels (coal gas and natural gas) were done in actual homes in Beijing.

2.1. Sampling protocol

Since it is known that emissions from most solid fuels can vary during the burning process (Smith et al., 1993; Cooper and Malek, 1982), integrated sampling is needed to cover a whole burn cycle (from fire start to fire extinction) in order to obtain emission data that can represent the real burning situation. In addition, as with many household appliances, it is necessary to choose a particular use cycle for fair comparison among different stoves. In the present study, we used the “water boiling test” procedure developed as a standard international method (VITA, 1985), i.e., a pot containing known amount of water was placed on a tested cookstove during each experiment. The initial and final water temperature and the amount of water evaporated was measured for each burn cycle. Hence, the energy received by the

pot can be determined. This procedure has an added advantage of enabling the simultaneous measurement of emissions and stove efficiencies, thus facilitating future calculations of the impact of changes in one or the other. Except for coal burning, the burn cycles ranged from 30 to 45 min for all other types of fuel/stove combinations. Coal burning needed a longer cycle (up to 6 h).

A typical sampling configuration from up-stream to down-stream included a sampling probe, a filter holder, a pump, and a Tedlar bag. For those having no flue, the stoves were placed under a hood built for the study, and the probe was placed inside of hood exhaust pipe. For those stoves with flues, the sampling probe was placed inside of the flue or inside of a hood which was placed over the end of the flue. Filters employed to collect total suspended particles (TSP) were heat-treated 37 mm quartz fiber filters (Pallflex Products, Putnam, CT). The sampling flow rate was adjusted to fill one or two 80-liter Tedlar bags throughout a burn cycle.

All 80-liter Tedlar bags were flushed at least three times with clean air before use. The bags were sealed at the end of the sampling period. Air samples were then taken from the 80-liter bags using glass syringes and either injected into a gas chromatograph (GC) for analysis or transferred in smaller (4-liter) Tedlar bags when local transportation of the air samples from burn test sites to the analytical laboratories were necessary. In addition, stainless-steel canisters were used to sample a subset of samples of the 80-liter Tedlar bags. The procedure was to fill an evacuated 0.85-liter canister to 2 atmospheres using a battery-operated pump, a process taking about 2 min. All Tedlar bag samples were stored less than 48 h before the GC analysis. The canister samples, however, were stored for up to 90 days due to complicated international shipping procedures.

Three successful tests with complete burn cycles were conducted for each fuel/stove combination. The flue gases collected in all three tests were measured in local laboratories of China and India for CO₂, CO, CH₄, and TNMHC. At least one background indoor air sample was taken for each fuel/stove combination and several outdoor samples were collected as well.

Table 1
CO emission ratios and emission factors for various biomass fuel/stove combinations

Fuel type	Fuel	Stove	Flue	Country	E_f (to CO ₂)		E_f (g/kg)		$E_{f,1}$ (g/task)	
					mean	c.v.	mean	c.v.	mean	c.v.
Dung cake	Cow dung cake	Hara	No	India	1.01E-1	0.30	6.1E+1	0.22	6.5E+1	0.32
	Cow dung cake	Traditional mud	No	India	7.60E-2	0.09	4.9E+1	0.08	4.4E+1	0.11
	Cow dung cake	Two-pot mud	Yes	India	4.50E-2	0.31	3.0E+1	0.26	2.6E+1	0.29
	Cow dung cake	Two-pot ceramic	Yes	India	4.80E-2	0.39	3.1E+1	0.35	2.1E+1	0.43
Crop residue	Wheat straw	Brick	Yes	China	7.36E-2	0.33	6.2E+1	0.27	4.8E+1	0.38
	Wheat straw	Improved brick	Yes	China	3.02E-1	0.31	1.7E+2	0.19	8.0E+1	0.27
	Maize stalk	Brick	Yes	China	5.51E-2	0.56	3.6E+1	0.49	2.2E+1	0.68
	Maize stalk	Improved brick	Yes	China	1.32E-1	0.39	7.9E+1	0.38	2.6E+1	0.42
	Mustard stalk	Traditional mud	No	India	8.00E-2	0.33	6.6E+1	0.30	3.2E+1	0.38
	Mustard stalk	Metal	No	India	6.60E-2	0.39	5.6E+1	0.35	1.6E+1	0.41
	Mustard stalk	Two-pot mud	Yes	India	1.38E-1	0.26	9.4E+1	0.28	4.2E+1	0.25
	Mustard stalk	Two-pot ceramic	Yes	India	7.70E-2	0.31	5.5E+1	0.27	1.8E+1	0.31
	Paddy straw	Traditional mud	No	India	7.00E-2	0.32	4.8E+1	0.30	3.7E+1	0.22
	Paddy straw	Two-pot mud	Yes	India	1.66E-1	0.64	1.0E+2	0.57	7.5E+1	0.66
Root fuel	Root fuel	Traditional mud	No	India	4.61E-2	0.47	4.5E+1	0.44	2.1E+1	0.52
	Root fuel	Metal	No	India	6.94E-2	0.23	6.7E+1	0.23	1.9E+1	0.29
	Root fuel	Two-pot mud	Yes	India	6.81E-2	0.14	5.5E+1	0.19	2.0E+1	0.13
Wood	Brush wood	Brick	Yes	China	1.20E-1	0.26	1.1E+2	0.21	5.2E+1	0.31
	Brush wood	Improved brick	Yes	China	1.88E-1	na	1.7E+2	na	7.6E+1	na
	Brush wood	Metal	No	India ^a	1.79E-1	0.16	1.6E+2	0.14	6.0E+1	0.15
	Fuel wood	Brick	Yes	China	4.07E-2	0.19	4.2E+1	0.17	1.9E+1	0.09
	Fuel wood	Improved brick	Yes	China	1.19E-1	0.54	9.9E+1	0.52	2.5E+1	0.59
	Fuel wood	Metal	No	India ^a	8.85E-2	0.31	7.4E+1	0.15	2.3E+1	0.55

Table 1 (Continued)

Fuel type	Fuel	Stove	Flue	Country	E_r (to CO ₂)		E_f (g/kg)		$E_{f,1}$ (g/task)	
					mean	c.v.	mean	c.v.	mean	c.v.
	Eucalyptus	Three-rock	No	India	6.17E-2	0.18	4.9E+1	0.17	1.8E+1	0.15
	Eucalyptus	Metal	No	India	6.82E-2	0.21	5.4E+1	0.23	1.7E+1	0.30
	Eucalyptus	Two-pot mud	Yes	India	1.64E-1	0.07	8.9E+1	0.02	2.7E+1	0.10
	Eucalyptus	Two-pot ceramic	Yes	India	9.41E-2	0.43	6.3E+1	0.41	1.4E+1	0.38
	Acacia	Three-rock	No	India	7.44E-2	0.07	5.0E+1	0.14	1.8E+1	0.14
	Acacia	Traditional mud	No	India	7.55E-2	0.16	4.8E+1	0.09	1.7E+1	0.12
	Acacia	Metal	No	India	7.31E-2	0.09	5.0E+1	0.19	1.3E+1	0.28
	Acacia	Two-pot mud	Yes	India	1.58E-1	0.07	7.5E+1	0.15	2.1E+1	0.07
	Acacia	Two-pot ceramic	Yes	India	9.24E-2	0.31	4.5E+1	0.29	1.0E+1	0.32
Charcoal	Charbriquette ^b	Angethi	No	India	1.20E-1	0.13	1.2E+2	0.12	6.6E+1	0.16
Charcoal	Charcoal	Angethi	No	India	1.80E-1	0.18	2.8E+2	0.15	6.2E+1	0.12

c.v. = coefficient of variation = (standard deviation)/mean; na: only one measurement was made.

^aThe stove was made in India but the test was conducted in China.

^bThe by-product left in gasifiers by which producer gas was made.

Table 2
CO emission ratios and emission factors for various fossil fuel/stove combinations

Fuel type	Fuel	Stove	Flue	Country	E_r (to CO ₂)		E_r (g/kg)		$E_{r,t}$ (g/task)		
					mean	c.v.	mean	c.v.	mean	c.v.	
Coal	Coal powder	Brick	Yes	China	1.21E-1	0.34	1.7E+2	0.39	4.4E+1	0.63	
	Coal powder	Metal	No	China	4.43E-2	na	7.1E+1	na	1.8E+1	na	
	Coal powder	Metal	Yes	China	5.64E-2	0.42	1.0E+2	0.31	3.1E+1	0.55	
	Washed coal powder	Metal	Yes	China	5.91E-2	0.15	8.6E+1	0.20	3.1E+1	0.25	
	Coal cake	Metal	No	China	6.59E-2	na	8.6E+1	0.02	6.0E+1	0.02	
	Honeycomb briquette S ^a	Metal	Yes	China	4.51E-2	0.38	6.7E+1	0.36	7.4E+1	0.39	
	Honeycomb briquette	Metal	No	China	4.19E-2	0.41	6.6E+1	0.35	1.5E+1	0.32	
	Honeycomb briquette	Metal	Yes	China	3.65E-2	0.21	6.0E+1	0.31	1.7E+1	0.30	
	Honeycomb briquette	Improved metal	No	China	3.44E-2	0.61	6.8E+1	0.50	7.6E-0	0.49	
	Coal briquette	Metal	Yes	China	2.32E-2	1.23	1.8E+1	0.34	4.9E-0	0.43	
	Coal briquette	Metal	No	China	1.96E-2	0.71	2.0E+1	0.72	3.9E-0	0.74	
	Kerosene	Kerosene	Wick	No	China	4.78E-3	0.21	8.2E-0	0.25	4.3E-1	0.19
		Kerosene	Wick	No	India	9.00E-3	0.22	1.8E+1	0.24	7.8E-1	0.29
Kerosene		Pressure	No	China	3.78E-3	0.48	6.6E-0	0.56	3.3E-1	0.56	
Kerosene		Pressure	No	India	3.30E-2	0.17	6.2E+1	0.17	3.0E-0	0.15	
LPG	LPG	Traditional	No	China	5.68E-3	0.74	2.1E-0	1.73	8.8E-2	1.72	
	LPG	Infrared head	No	China	9.64E-3	0.60	1.7E+1	0.68	8.8E-1	0.89	
	LPG	LPG burner	No	India	7.00E-3	0.21	1.5E+1	0.21	6.1E-1	0.21	
Gas	Natural gas	Traditional	No	China	1.20E-4	1.73	3.9E-1	1.26	1.4E-2	1.20	
	Natural gas	Infrared head	No	China	3.86E-4	1.81	5.8E-1	1.81	5.3E-3	1.81	
	Coal gas	Traditional	No	China	4.91E-5	2.00	3.0E-2	2.00	1.4E-3	2.00	
	Biogas ^b	Biogas stove	No	India	2.00E-3	0.50	2.0E-0	0.50	1.9E-1	0.39	

c.v. = coefficient of variation = (standard deviation)/mean; na: only one measurement was made.

^a From a special coal mine in Shanxi province.

^b Biogas, or producer gas, is produced from dung and other biowaste, so it may not be considered as a fossil fuel.

2.2. Analytical methods

In both China and India, GCs were set up to analyze background samples and samples taken out of the filled Tedlar bags for CO₂, CO, CH₄, and TNMHC. A system of GC-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 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 using a FID and a blank GC column (the air peak was corrected). All GCs were calibrated daily with locally made standards and periodically checked with a standard gas mixture of CO₂, CO, CH₄ prepared by Scott Specialty Gases, USA. The agreement between the locally made standards and US made standards was $< \pm 4\%$.

The filled canisters were shipped back to Oregon Graduate Institute of Science and Technology (OGIST) to be analyzed mainly for hydrocarbon speciation. These canister samples were also analyzed for CO₂, CO, CH₄, and TNMHC, providing data for inter-laboratory comparison.

Fuel and ash analyses (calorific values, carbon contents, sulfur contents, moisture contents, etc.) were completed locally by the standard fuel analysis methods. Carbon contents of TSP collected on quartz fiber filters were measured using a thermal-optical carbon analysis technique (Johnson et al., 1981) at Sunset Laboratory, Oregon, U.S.A.

2.3. Quality assurance

The same sampling sheets and data input methods were used in China and India to reduce the confusion when handling and transferring data. Similar sampling and analytical procedures were followed by both laboratories.

Prior to the three planned tests for each fuel/stove combination, trial runs were conducted until a satisfactory method precision was obtained. At least one set of parallel sampling was conducted for each fuel type. Results from these replicate

samples were $< 20\%$ RSD. 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 $R^2 > 0.99$.

Inter-laboratory checks were also conducted. The three laboratories were provided with a standard mixture of CO₂, CO, and CH₄ from the same source (Scott Specialty Gases, U.S.A.). Results obtained by the local GC analyses were compared with results of canister samples analyzed by OGIST (see Fig. 1). The China results and OGIST results appeared to be in fairly good agreement: the line in the top panel of Fig. 1 is a 1:1 line. The India results, however, were systematically lower than the OGIST data: the line in the bottom panel of Fig. 1 is a linear regression line with a $R^2 = 0.93$. Based upon the comparison between the China and OGIST results and our belief that the OGIST lab has extensive experiences in GC analysis, we have corrected all GC results of the India lab using linear regression equations derived from split

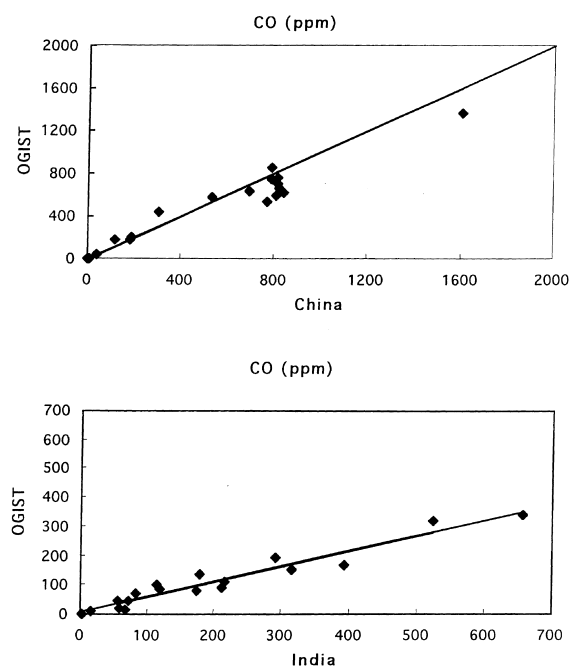


Fig. 1. Comparison of results from the OGIST lab and the labs in China and India. (Note: the results are derived from analyses of split samples.)

sample analyses by the two labs (in Fig. 1, [OGIST data] = 8.32 + 0.52 [India data]). The corrected values were reported and used for emission factor calculations.

2.4. Carbon balance model

As used in the Manila pilot study (Smith et al., 1993), a carbon balance model was used in the present study to calculate the emission factors for each fuel/stove combination tested. The mass balance for carbon can be described as follows,

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

where C_f = carbon mass in the fuel, C_a 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 in some of the solid fuels. In this case, the carbon mass of the lighter fluid was added to the left-hand side of the Eq. 1. Rearranging Eq. 1 leads to:

$$\frac{C_f - C_a}{C_{CO_2}} = 1 + \frac{C_{CO}}{C_{CO_2}} + \frac{C_{CH_4}}{C_{CO_2}} + \frac{C_{TNMHC}}{C_{CO_2}} + \frac{C_{TSP}}{C_{CO_2}} = 1 + K \quad (2)$$

because,

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

where $\frac{C_{CO}}{C_{CO_2}}, \frac{C_{CH_4}}{C_{CO_2}}, \frac{C_{TNMHC}}{C_{CO_2}}, \frac{C_{TSP}}{C_{CO_2}}$ are called the emission ratios of CO, CH_4 , TNMHC, and TSP carbon, 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. Thus,

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

If the emission factor (E_f) for a pollutant is defined as mass of the pollutant emitted per mass

of fuel burned (M), the emission factor for CO_2 can be calculated from the following equations

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

(unit : carbon mass in CO_2 per kg of fuel)

or

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

(unit : CO_2 mass per kg of fuel)

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 the other pollutants can now be calculated easily from the CO_2 emission factor and their emission ratios to CO_2 . For example,

$$CO E_{f,C} = CO_2 E_{f,C} \frac{C_{CO}}{C_{CO_2}}, \quad (7)$$

(unit : carbon mass in CO per kg of fuel)

or

$$CO E_f = CO E_{f,C} \times 2.33 \quad (8)$$

(unit : CO mass per kg of fuel).

One of the advantages of using this carbon balance model to determine emission factors is that this method 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 the species 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 $E_{f,t}$ (mass of pollutant per cooking task) rather than the fuel mass based are a better performance index to compare the air pollution potential of different fuel/stove combinations (Joshi et al., 1989; Zhang and Smith, 1996a). The simplest task measure is the release per unit energy delivered to the pot (g/kJ).

The following two parameters are needed to convert emissions per kg of fuel to emissions per task: fuel calorific value (H in kJ/kg) and stove efficiency (η in %),

$$\text{CO}E_{f,t} = \frac{\text{CO}E_f}{H\eta}J, \quad (9)$$

(unit : CO mass per task)

where J = energy delivered per standard task (kJ/task). In this paper, the standard cooking task was defined as one that would deliver 1 MJ heat to the pot (modified from Ahuja et al., 1987 and Joshi et al., 1989). H and η were measured in the study and reported elsewhere (Smith et al., 1999).

3. Results and discussion

3.1. Molar emission ratios

Under a hypothetical condition for a perfectly complete combustion, all the carbon contained in the fuel would turn into CO_2 . Therefore, compounds other than CO_2 found in the flue gas are typically called products of incomplete combustion (PIC); the emission ratio of total PIC to CO_2 usually serves as an indicator of combustion efficiency (Smith et al., 1999); and the emission ratio of an individual PIC is often used as a relative parameter for the source strength of the PIC (Smith et al., 1993; Crutzen and Andreae, 1990). Because sampling positions in the flue gas stream were not identical for different burns, CO concentration values alone are difficult to interpret. Instead, since CO_2 is a major product emitted from fuel combustion and dilution factors for CO_2 , CO and other species can be considered equal (Smith et al., 1993), CO emission ratios (E_r) are presented in Tables 1 and 2. Calculation of E_r was based on the following equation:

$$\text{CO}E_r = \frac{(\text{CO})_f - (\text{CO})_b}{(\text{CO}_2)_f - (\text{CO}_2)_b}, \quad (10)$$

where $(\text{CO})_f$ = CO concentration in flue gases (ppb), $(\text{CO})_b$ = background CO concentration (ppb), $(\text{CO}_2)_f$ = CO_2 concentration in flue gases (ppb), and $(\text{CO}_2)_b$ = background CO_2 concentra-

tion (ppb). The emission ratios calculated from Eq. 10 using molar (or equivalent) concentrations are virtually the same as those defined in Eq. 3 based on carbon mass.

Reported in Tables 1 and 2 are mean values and coefficients of variation (c.v.) from three tests for each fuel/stove combination. In general, results from the three tests agreed well (with a low c.v.). The c.v. values for gas/stove tests tended to be larger. This is because these stoves emitted CO concentrations close to background CO concentration and the analytical detection limit.

Across the 56 fuel/stove combinations tested, $\text{CO}E_r$ ranged widely from 4.91×10^{-5} for the coal gas/traditional stove to 3.02×10^{-1} for the improved brick stove burning wheat straw. The range of $\text{CO}E_r$ for all the biomass fuel/stove combinations and the coal/stove combinations, however, was a little over an order of magnitude: from 1.96×10^{-2} for the coal briquette/metal stove to 3.02×10^{-1} for the wheat straw/improved brick. Except for the kerosene pressure stove tested in India, CO emission factors for the other two kerosene stoves, the LPG stoves, and the biogas stoves, were about 1 to 2 orders of magnitude lower than for all the tested biomass and coal stoves. Burning natural gas and coal appeared to generate the lowest $\text{CO}E_r$.

3.2. Emission factors

Fuel mass based and task based CO emission factors are listed in Tables 1 and 2. The results indicate that:

1. CO emission factors were dependent upon not only fuel types, but also stove types. For example, the range of fuel mass based $\text{CO}E_f$ (g/kg) for the following fuel/stove combinations was close to an order of magnitude: 10 combinations of crop residue/stove, 15 wood/stove, 11 coal/stove, and 4 kerosene/stove (see Table 3).

2. Across the 56 fuel/stove combinations tested, CO emission factors ranged widely. For example, the fuel mass based $\text{CO}E_f$ ranged from 3.0×10^{-2} g/kg for the coal gas/traditional stove to 2.8×10^2 g/kg for the charcoal/Angethi stove, nearly 4 orders of magnitude. The task based $\text{CO}E_{f,t}$ ranged from 1.4×10^{-3} g/task for coal gas/traditional stove

Table 3

CO emission factors (E_f) for various biomass and fossil fuels (when they are burned in cookstoves)

Fuel type	Number of fuel/stove types	E_f (g/kg)			E_f (g/task)		
		Mean	Min.	Max.	Mean	Min.	Max.
Dung cake	4	4.3E+1	3.0E+1	6.1E+1	3.9E+1	2.1E+1	6.5E+1
Crop residue	10	7.7E+1	3.6E+1	1.7E+2	4.0E+1	1.6E+1	8.0E+1
Root fuel	3	5.6E+1	4.5E+1	6.7E+1	2.0E+1	1.9E+1	2.1E+1
Wood	15	7.8E+1	4.2E+1	1.7E+2	1.7E+1	3.2E-0	4.4E+1
Charcoal	2	2.0E+2	1.2E+2	2.8E+2	2.7E+1	1.0E+1	7.6E+1
Coal	11	7.4E+1	1.8E+1	1.7E+2	2.8E+1	3.9E-0	7.4E+1
Kerosene	4	2.4E+1	6.6E-0	6.2E+1	1.1E-0	3.3E-1	3.0E-0
LPG	3	1.1E-0	2.1E-0	1.7E+1	5.3E-1	8.8E-2	8.8E-1
Biogas	1	2.0E-0	2.0E-0	2.0E-0	1.9E-1	1.9E-1	1.9E-1
Natural gas	2	4.9E-1	3.9E-1	5.8E-1	9.7E-3	5.3E-3	1.4E-2
Coal gas	1	3.0E-2	3.0E-2	3.0E-2	1.4E-3	1.4E-3	1.4E-3

to 0.8×10^2 g/task for wheat straw/improved brick stove, almost 5 orders of magnitude. Although the charcoal/Angethi had the highest fuel mass based CO E_f , the highest task based CO $E_{f,t}$ was for the wheat straw/improved brick stove. This is because charcoal has a higher carbon content (and thus energy content) than crop residues and that the charcoal stove may have a higher energy efficiency. Even though burning a kg of charcoal produces more CO than burning a kg of wheat straw, performing the same cooking task would emit less CO.

3. Among the fossil fuels tested, coal had the highest CO E_f . The CO E_f for coal was comparable to those for biomass fuels. Performing the same cooking task using any gas fuels, LPG, and kerosene would emit significantly less CO than using biomass and coal.

4. Improved stoves did not necessarily have lower CO emission factors compared to the traditional stoves of the same type. In fact, it is often true that improved stoves tend to generate more PICs (Zhang and Smith, 1996b). We also found this being true in the present study for CO emission factors when comparing the traditional brick stove vs. the improved brick stove regardless of wheat straw, maize stalk, brush wood, or fuel wood. Although improved stoves often increase overall fuel efficiency, which is the product of combustion efficiency and heat transfer efficiency,

through increasing the heat transfer efficiency, in many cases they decrease the combustion efficiency (Smith, 1987). A decreased combustion efficiency could lead to less complete combustion which emits more PICs including CO.

3.3. CO emissions from combustion of household fuels

The emission factors reported in this paper would enable an accurate estimate on CO emissions from combustion of domestic fuels if stove-specific fuel consumption data are available. Unfortunately, the regional or national fuel consumption data have often not been stove-specific, perhaps due to the lack of such a detailed database of emission factors. We thus hope that these fuel/stove specific emission factors stimulate the collection of stove-specific fuel data in the future.

To obtain a rough idea about the relative importance of CO emissions from household fuel usage, we estimated CO emissions from major domestic fuels in China and India using the CO E_f measured in this study and the fuel consumption data "best" available to us. The results are shown in Table 4 for China and Table 5 for India. For China, the fuel consumption data was for 1994 and was obtained from the Institute for Techno-Economics and Energy System Analysis (ITESSA) of Tsinghua University, Beijing, China; the CO E_f

Table 4
Estimated CO emissions from major household fuels in China in 1994

Fuel type	Fuel consumed (Tg/yr) ^a	CO E_f used (g/kg)	CO emitted (Tg/yr)
Dung	3.5		0.15
Crop residue	283	77	21.8
Wood	163.6	78	12.8
Total biomass	450.1		34.75
Coal	175.7	74	13.0
Kerosene	1.09	24	0.026
LPG	3.85	1.1	0.0042
Natural gas	1.74 (2E+9 m ³ /yr) ^b		0.00085
Coal gas	2.34 (5.4E+9 m ³ /yr) ^b	0.03	0.000070
Total fossil	184.7		13.03

^a Source: ITEESA, Tsinghua University, Beijing, China.

^b Density of natural gas = 0.868 kg/m³, density of coal gas = 0.433 kg/m³.

Table 5
Estimated CO emissions from major fuel/stove combinations in India (1990–1991)

Fuel/stove	Fuel consumed (Tg/yr) ^a	CO E_f used (g/kg) ^b	CO emitted (Tg/yr)
Dung/traditional mud	31.6	49	
Dung/improved mud		30	
Dung/Hara	19.12	61	1.17
Crop residue/trad. mud.	58.6	65	3.81
Crop residue/improved mud	2.9	90	0.261
Crop residue/improved metal	0.3	56	0.017
Wood/traditional mud		48	8.83
Wood/improved mud		122	0.775
Wood/improved metal		58	0.041
Charcoal/Angethi	0.5	275	0.138
Total biomass	316.3		16.68
Coal	6	74	0.44
Kerosene/wick	2.16	18	0.039
Kerosen/pressure	1.82	62	0.113
LPG	2.1	15	0.032
Biogas	0.5 (6.66E+8m ³)	2	0.001
Total fossil	12.6		0.63

^a Source: Smith et al., 1999.

^b Weighted averages based upon fuel and stove distributions (Smith et al., 1999).

used were simply the averages for each fuel type, as shown in Table 3, due to the lack of the stove-specific data. In Table 4, the amount of coal consumed was for both cooking and heating (the stoves were, in most cases, the same for cooking

and heating), while other types of fuels were mainly for cooking. For India, since more detailed information on stove and fuel distributions were known, the CO E_f used for calculating CO emissions were weighted averages (Smith et al., 1999).

Since none of coal stoves was tested in India, the average CO E_f measured in China was used to calculate CO emissions from coal combustion in India cookstoves.

Our rough estimates show that China emitted 47.78 Tg/yr of CO from combusting 634.8 Tg/yr of fuels in residential sector in 1994 and that India emitted 17.31 Tg/yr of CO from combusting 328.9 Tg/yr of household fuels in 1990–991. In India, fossil fuels contributed about 4% to total household fuels in terms of fuel mass; they contributed about 4% to the total CO emission from all household fuels. In China, fossil fuels, in which 95% was coal, accounted for 29% of total household fuels. Due to the large fraction of coal in fossil fuels and high CO E_f for coal, combustion of fossil fuels accounted for 27% of the total CO emission from all household fuels in China.

Globally, it was estimated that combustion of fossil fuels releases 500 Tg/yr of CO and that biomass combustion (both anthropogenically and naturally) emits 680 Tg/yr of CO (Khalil and Rasmussen, 1990). Based upon the estimates shown in Tables 4 and 5, biomass combustion in cookstoves in China and India alone emits 51.43 Tg/yr of CO, or 8% of the total biomass emission. On the other hand, CO emitted from burning fossil fuels in cookstoves in India and China together accounts for about 3% of the total global fossil fuel emission, a relatively small amount, although perhaps larger than the fraction of total fossil fuel consumption represented by this use.

4. Conclusions and implications

This paper presents a new and comprehensive database of CO emission factors for 56 fuel/stove combinations commonly used in developing countries. These include various stoves (e.g., traditional, improved, mud, brick, and metal, with and without chimney) using animal dung, different species of crop residues and wood, root fuel, charcoal, kerosene, and several types of coals and gases. The chosen fuel/stove combinations represent a large fraction of the total in developing countries and a large fraction of biomass stoves

used worldwide. Thus the database can be used to improve estimates of national and global CO emission inventories.

The CO emission factors measured were not only a function of fuel type, but also of stove type. This suggests that the emission factors measured under open or large-scale combustion conditions (e.g., open fire of biomass, industrial scale burning of coal or other fuels) would not be applicable for the enclosed or small-scale combustion devices such as cookstoves.

Across the fuel/stove combinations tested, CO emission factors ranged widely. In general, biomass fuels and coal had higher CO emission factors than other fossil fuels. Due to the large biomass consumption and the high biomass emission factors, a rough estimate, based on fuel consumption data of China and India, indicates that biomass combustion in household cookstoves may contribute significantly to the total CO emission from all sources of biomass combustion globally.

Simultaneous measurement of stove energy efficiencies and fuel energy contents enabled us to report CO emission factors on the basis of a standard cooking task as well. These data will be further analyzed to assess impacts on CO emissions and exposures of changes brought about by fuel switching, stove switching, or improvement, and other technical and policy options, as discussed in Part 2 of our report on CO emissions from these various cookstoves (see this same issue of *Chemosphere*).

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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.
- 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. *Atmos. Environ.* 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.
- Delmas, R., Lacaux, J.P., Brocard, D., 1995. Determination of biomass burning emission factors: methods and results. *Environ. Monitoring Assessment* 38, 181–204.
- FAO, Food and Agriculture Organization, 1987. Yearbook of Forest Products. Rome.
- Hubbard, A.J., 1995. Hazardous air emissions potential from a wood-fired furnace. *Combustion Sci. Technol.* 108, 297–309.
- Hurst, D.E., Griffinth, D.W.T., Carras, J.N., Williams, D.J., Fraser, P.J., 1994a. Measurements of trace gases emitted by Australian savanna fires during the 1990 dry season. *J. Atmos. Chem.* 18, 33–56.
- Hurst, D.E., Griffinth, D.W.T., Cook, G.D., 1994b. Trace gas emissions from biomass burning in tropical Australian savannas. *J. Geophys. Res.* 99, 16441–16456.
- IPCC 1990. Climate Change: The IPCC Scientific Assessment, Intergovernmental Panel on Climate Change, Cambridge Univ. Press, Cambridge.
- 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. *Agri. Ecosys. Environ.* 38, 313–330.
- Johnson, R.L., Shah, J.J., Cary, R.A., Huntzicher, J.J. 1981. An automated thermal-optical method for the analysis of carbonaceous aerosol In: Macias, E.S., Hopke, P.K. (Eds.) ACS Symposium Series No. 167, Atmospheric Aerosol: Source/Air Quality Relationships.
- Joshi, V., Venkataraman, C., Ahuja, D.R., 1989. Emissions from burning biofuels in metal cookstoves. *Environ. Management* 13, 763–772.
- Khalil, M.A.K., Rasmussen, R.A., 1985. Causes on increasing atmospheric methane: depletion of hydroxyl radicals and the rise of emissions. *Atmos. Environ.* 19, 397–407.
- Khalil, M.A.K., Rasmussen, R.A., 1990. The global cycle of carbon monoxide: trends and mass balance. *Chemosphere* 20, 227–242.
- Klouda, G.A., Connolly, M.V., 1995. Radiocarbon (^{14}C) measurements to quantify sources of atmospheric carbon monoxide in urban air. *Atmos. Environ.* 29, 3309–3318.
- Seinfeld, J.H., Pandis, S.N., 1997. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. Wiley/Interscience, New York.
- Smith, K.R., 1987. Biofuels, Air Pollution, and Health. Plenum Press, New York.
- Smith, K.R., Zhang, J., Thorneloe, S.A. 1995. Greenhouse gases from widely used small-scale combustion devices in developing countries: Phase I-II: stoves in India and China and Phase III: charcoal kilns in Thailand. In: Proceedings of the EPA's Symposium on Greenhouse Gas Emissions and Mitigation Research. 27–29 June, 1995, Washington, DC.
- 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.
- 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., Washington, D.C., 1999. Greenhouse Gases from Small-scale Combustion Devices in Developing Countries, Phase IIa: Household Stoves in India. Submitted for EPA review and publication, Office of R&D, US EPA.
- VITA, Volunteers in Technical Assistance Inc., (1985). Testing The Efficiency of Wood-burning Cookstoves: International Standards, Arlington, VA.
- Ward, D.E., Hardy, C.C., 1991. Smoke emissions from wildland fires. *Environ. Internat.* 17, 117–134.
- Zhang, J., Smith, K.R., 1996a. Hydrocarbon emissions and health risks from cookstoves in developing countries. *J. Exposure Analysis Environ. Epidemiol.* 6, 147–161.
- Zhang, J., Smith, K.R., 1996b. Indoor air pollution: Formaldehyde and other carbonyls emitted from various cookstoves. In: Proceedings of Indoor Air '96, the Seventh International Conference on Indoor Air Quality and Climate, Vol. 2, pp. 85–90. Nagoya, Japan.
- Zhang, J., Smith, K.R., Kishore, V.V.N., Ma, Y., Rasmussen, R., Uma, R., Khalil, M.A.K., Kusam, J., Thorneloe, S.T. 1997. Greenhouse Gases from Cookstoves in developing countries: preliminary emission factors. In: Emission Inventory: Planning for the Future, Proceedings of the 1997 AWMA/EPA Emission Inventory Conference, Vol. 1, pp. 368–378. Research Triangle Park, NC.

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