New methodology for estimating biofuel consumption for cooking: Atmospheric emissions of black carbon and sulfur dioxide from India

Gazala Habib,[1] Chandra Venkataraman,[1] Manish Shrivastava,[2] Rangan Banerjee,[3] J. W. Stehr,[4] and Russell R. Dickerson[4]

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[1] The dominance of biofuel combustion emissions in the Indian region, and the inherently large uncertainty in biofuel use estimates based on cooking energy surveys, prompted the current work, which develops a new methodology for estimating biofuel consumption for cooking. This is based on food consumption statistics, and the specific energy for food cooking. Estimated biofuel consumption in India was 379 (247–584) Tg yr^{-1}. New information on the user population of different biofuels was compiled at a state level, to derive the biofuel mix, which varied regionally and was 74:16:10%, respectively, of fuelwood, dung cake and crop waste, at a national level. Importantly, the uncertainty in biofuel use from quantitative error assessment using the new methodology is around 50%, giving a narrower bound than in previous works. From this new activity data and currently used black carbon emission factors, the black carbon (BC) emissions from biofuel combustion were estimated as 220 (65–760) Gg yr^{-1}. The largest BC emissions were from fuelwood (75%), with lower contributions from dung cake (16%) and crop waste (9%). The uncertainty of 245% in the BC emissions estimate is now governed by the large spread in BC emission factors from biofuel combustion (122%), implying the need for reducing this uncertainty through measurements. Emission factors of SO_2 from combustion of biofuels widely used in India were measured, and ranged 0.03–0.08 g kg^{-1} from combustion of two wood species, 0.05–0.20 g kg^{-1} from 10 crop waste types, and 0.88 g kg^{-1} from dung cake, significantly lower than currently used emission factors for wood and crop waste. Estimated SO_2 emissions from biofuels of 75 (36–160) Gg yr^{-1} were about a factor of 3 lower than that in recent studies, with a large contribution from dung cake (73%), followed by fuelwood (21%) and crop waste (6%). *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; *KEYWORDS:* aerosols, emission inventory, regional pollution

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1. Introduction

[2] Biomass burning is a significant source of emissions in the Indian region, as evidenced by large concentrations of tracers like acetonitrile and particulate potassium, and the attribution of carbon monoxide primarily to this source during the INDOEX campaign [*Reiner et al.*, 2001; *de Laat et al.*, 2001; *Ball et al.*, 2003]. Important constituents of fine aerosol (dp < 1 μm) measured over the Indian region included black carbon (BC) and sulfate contributing about 14% and 32% of dry mass, respectively [*Ramanathan et al.*, 2001; *Neusüß et al.*, 2002; *Ball et al.*, 2003].

[3] The contribution of biomass burning to aerosol emissions in the Indian region, especially BC, is under debate. During INDOEX it was suggested that fossil fuel burning was the dominant source of BC, based on a low organic carbon/BC and low sulfate/BC ratio in aerosols measured over the Indian Ocean [*Novakov et al.*, 2000; *Lelieveld et al.*, 2001; *Ramanathan et al.*, 2001; *Mayol-Bracero et al.*, 2002]. However, another finding [*Guazzotti et al.*, 2003] was the large predominance of submicron, chemically mixed particles, containing carbon and potassium. This

[1]Department of Chemical Engineering, Indian Institute of Technology Bombay, Powai, Mumbai, India.
[2]Center for Environmental Science and Engineering, Indian Institute of Technology Bombay, Powai, Mumbai, India.
[3]Energy Systems Engineering, Indian Institute of Technology Bombay, Powai, Mumbai, India.
[4]Department of Meteorology, University of Maryland, College Park, Maryland, USA.

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mixing with potassium, the established tracer for biomass burning [*Chow et al.*, 1994; *Wei et al.*, 1996], in source apportionment methods [*Miller et al.*, 1972; *U.S. Environmental Protection Agency* (*EPA*), 1989], implies the potential origin of BC in the Indian region from biomass burning, with potential contributions from the Indian subcontinent, Southeast Asia, and Africa.

[4] It has recently been suggested that biomass burning in India is dominated by biofuel combustion (i.e., burning of fuelwood, dung cake, and crop waste in cooking stoves) in contrast to global biomass burning, which is dominated by open burning of crop waste and forest biomass [*Reddy and Venkataraman*, 2002; *Streets et al.*, 2003; *Bond et al.*, 2004]. The emission source influences the composition of pollutant aerosols, particularly organic compounds, their water uptake, and consequent optical and radiative effects. Therefore this work focuses on improving emissions estimation from the regionally dominated source of biofuel combustion to assist potential improvement in regional climate change assessment and global climate prediction.

[5] Bottom-up emissions estimates from biofuel combustion [*Reddy and Venkataraman*, 2002; *Streets et al.*, 2003; *Yevich and Logan*, 2003; *Bond et al.*, 2004] have relied on energy-survey based activity data in which uncertainties in the per capita biofuel use were not well characterized. In addition, the user population for various biofuels was typically not reported, and was assumed [*Streets and Waldhoff*, 1998; *Reddy and Venkataraman*, 2002], leading to further uncertainties. Specifically for India, the per capita biofuel consumption [*Reddy and Venkataraman*, 2002] was derived from energy use surveys, for example, those carried out during 1985–1992 [*Sinha et al.*, 1998], in 15 agroclimatic zones of India. Sample sizes were small, leading to large uncertainty in the per capita biofuel use. In the absence of specific information, the entire rural population was assumed to use all three biofuels. In order to address these methodological drawbacks, one focus of the present work was the development of a new methodology for estimating biofuel use for cooking based on food consumption statistics and the specific energy requirement for food preparation.

[6] In addition, very few measurements have been reported for trace gas emission factors from biofuel combustion. For biofuels, emission inventories have arrived at best approximations of emission factors of sulfur dioxide from measurement of typical sulfur content of the fuel and assumed percentage retention of sulfur in ash [*Streets and Waldhoff*, 1998; *Olivier et al.*, 2001; *Reddy and Venkataraman*, 2002]. Recent studies of direct measurement of SO_2 emission factors from combustion of some biofuels [*Ballard-Tremeer and Jawurek*, 1996; *Zhang et al.*, 2000] gave widely varying results using different methods. Therefore another focus of present study was to fill the gap in SO_2 emission factors from biofuel combustion, with measurements for a comprehensive set of biofuels used in India.

[7] An important goal of this work was to reduce the uncertainty in regional emissions, which are an important input to climate change modeling and assessment, and to develop emissions for a recent base year. Importantly, the uncertainty in all input variables was established and propagated to generate 95% confidence intervals (CI) on estimates of biofuel use for cooking, and associated BC and sulfur dioxide emissions from India, for the base year of 2000.

2. Methodology for Estimating Energy/Fuel Consumption for Food Preparation

[8] Biofuels are currently estimated to supply about 85–90% of cooking energy consumption in rural India [*Tata Energy Research Institute*, 1995]. The method developed in this work for estimating energy/fuel consumption for cooking was based on food consumption statistics (F_{ij}, kg per capita per day) in each state i, for each of four cooking processes j available from *National Sample Surveys* (*NSS*) [2001] both for rural and urban regions (Figure 1). This was combined with the specific energy required for food preparation (EM_{jk}, MJ kg^{-1} of food cooked) for various cooking processes j, using different fuels k [*Verhaart*, 1982; *Islam et al.*, 1984; *Mukunda et al.*, 1988; *Ravindranath and Ramakrishna*, 1997].

[9] State population (P_i) combined with fraction fuel user population (f_{ik}%) in state i using various fuels k was derived separately for rural and urban regions from the *National Family Health Survey* (*NFHS*) [2001]. The end use energy for cooking (EE_{ijk}) (equation (1)), defined as the amount of energy required as input into the cooking process, is the product of the per capita food consumption, specific cooking energy, and state population using a particular fuel. This end use energy when divided by the cooking device efficiency (η_k) of each fuel type k gives the energy consumed (EC_{ik}) (equation (2)), and further divided by the fuel lower heat value (Q_k) gives the fuel consumption in mass units, which can be calculated on an annual basis (M_{ik}) (e.g., Tg yr^{-1} of fuelwood) in each state (equation (3)).

$$EE_{ijk} = F_{ij} \times EM_{jk} \times P_i \times f_{ik}, \qquad (1)$$

$$EC_{ik} = \left[\frac{\sum_{j=1}^{4} \left(EE_{ijk} \right)}{\eta_k} \right], \qquad (2)$$

$$M_{ik} = \left[\frac{\sum_{j=1}^{4} \left(F_{ij} \times EM_{jk} \times P_i \times f_{ik} \right)}{\eta_k \times Q_k} \right]. \qquad (3)$$

This new methodology allows us to derive bounds on the mean estimates of biofuel use by propagating the measured or estimated variance on the input parameters.

3. Food Consumption Statistics, Specific Energy, and Fuel Mix

[10] Food consumption statistics, available through National Sample Surveys conducted annually in 25 states

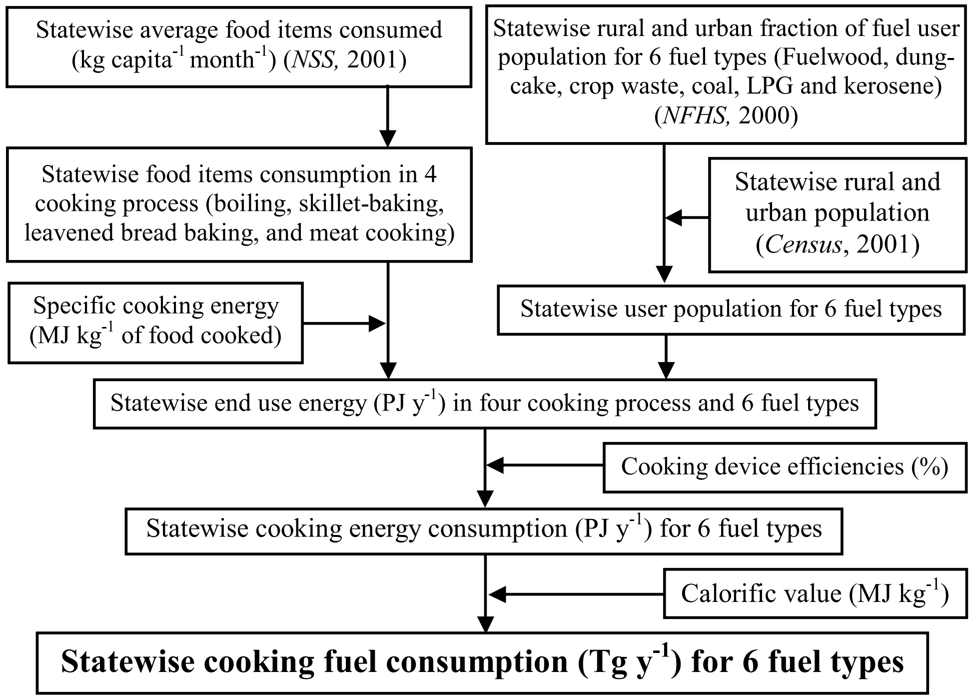

Figure 1. Methodology for estimation of statewise fuel use for cooking in India based on food consumption statistics, specific energy requirement for food preparation, and fuel user population.

and seven union territories, cover about 0.1% of the Indian population (about 1 million people), and have an uncertainty of about 3% [*Rao and Sharma*, 1980]. Statewise average food item consumption (kilograms per capita per day) for rural and urban regions for the year 1999–2000 [*NSS*, 2001] were aggregated according to four common cooking processes, i.e., boiling (cereals, pulses/lentils: the type of edible seeds peas and beans, vegetables, milk, tea, and eggs), skillet-baking (unleavened wheat-bread or "chapattis"), baking (leavened wheat-bread), and meat cooking (boiling plus frying). Estimated national food consumption was 332 Tg yr^{-1}, with higher per capita and total consumption in the northern, western, and eastern regions. Among food preparation processes, boiling contributed to the largest food consumption (74%), while meat cooking contributed to the least consumption (5%).

[11] The above food consumption statistics were combined with specific energy for food preparation, which was reported from the experiments in households or in the laboratory by measuring fuel used for various cooking tasks [*Islam et al.*, 1984; *Mukunda et al.*, 1988; *Ravindranath and Ramakrishna*, 1997]. These specific energies were compiled for the four cooking processes using six fuels, i.e., fuelwood, dung cake, crop waste, coal, LPG, and kerosene from reported measurements (Table 1). As there was no significant difference in specific energy for food preparation among the biofuels, these were aggregated (Table 1). The energy needed for rice boiling was based on a water to rice ratio of 6:1, most frequently used for rice cooking [*Ramesh and Rao*, 1996]. The reported energy for leavened bread baking and meat cooking [*Verhaart*, 1982] was theoretically determined.

[12] The *NFHS* [2001] reports statewise population, in rural and urban regions, using the six cooking fuels listed above, for 14 major states comprising 90% of national population. The rural population primarily uses biofuels (90% on average), while the urban population uses fossil fuels (73% on average) for cooking. Cooking-fuel use bears a relation to fuel accessibility, with dung cake and crop

Table 1. Specific Energy for Four Cooking Processes Using Various Fuels[a]

	Specific Energy for Cooking, MJ kg^{-1}			
Fuel Types	Boiling	Skillet Baking	Baking Leavened Bread	Meat Cooking
Biofuel	3.4 ± 0.3[b,c,d,e,f] (7, 9%)	2.4 ± 0.7[e] (4, 30%)	6.7[f]	4.1 ± 0.2[f] (3, 5%)
Coal	2.09[c] (1)	2.4 ± 0.7[e] (4, 30%)	6.7[f]	4.1 ± 0.2[f] (3, 5%)
LPG	1.9[c] (1)	1.2–2.0[e] (2)	6.7[f]	4.1 ± 0.2[f] (3, 5%)
Kerosene	3.5 ± 0.3[c,e] (4, 8%)	1.2–2.0[e] (2)	6.7[f]	4.1 ± 0.2 (3, 5%)

[a]Values in parentheses are number of experiments and the coefficient of variance.
[b]Mean and 1 standard deviation around mean.
[c]*Ravindranath and Ramakrishna* [1997].
[d]*Islam et al.* [1984].
[e]*Mukunda et al.* [1988].
[f]*Verhaart* [1982].

Table 2. Thermal Efficiency of Cooking Stoves and Calorific Value of Various Fuels[a]

Stove-Fuel Systems	Device Efficiency, %	Calorific Value, MJ kg^{-1}
Traditional stove/wood	13.8 ± 2.2[b,c,d,e,f,g,h] (17, 16%)	16.2 ± 1.7[c,d,e,f,g,h,i] (7, 11%)
Traditional stove/dung-cake	11.07 ± 2.0[c,d,e,f,g] (10, 19%)	11.8 ± 2.0[c,d,e,f,g] (4, 17%)
Traditional stove/crop waste	11.8 ± 3.0[c,d,f] (10, 25%)	15.2 ± 2.8[c,d,f,j] (19, 18%)
Angethi/char briquette	16.4[e] (1)	15.9[e] (1)
Pressure and wick/kerosene	49.4 ± 8.2[e,f,k] (4, 17%)	42.6 ± 1.5[e,f,k] (4, 3%)
LPG stove/LPG	57 ± 4.8[e,f,k] (3, 8%)	45.9 ± 0.1[e,f,k] (3, 0.2%)

[a]Values in parentheses are number of experiments and coefficient of variance.
[b]Mean and 1 standard deviation around mean.
[c]*Joshi et al.* [1989].
[d]*Kandpal and Maheshwari* [1995].
[e]*Ravindranath and Ramakrishna* [1997].
[f]*Smith et al.* [2000].
[g]*Venkataraman and Rao* [2001].
[h]*Gupta et al.* [1998].
[i]*Ahuja et al.* [1987].
[j]*Koopmans and Koppejan* [1997].
[k]*Kandpal et al.* [1995].

wastes being important sources of energy in large portions of the country where fuelwood is scarce [*Ravindranath and Ramakrishna*, 1997; *Banerjee et al.*, 1999]. This is reflected in the 10–20% rural user population of dung cake in northern and eastern regions and crop waste in eastern region where the fuelwood is scarce, compared to less than 1% in the west and south, the regions where fuelwood is easily available. Reported biofuel users in urban India are limited to 20% for wood, and less than 2% for dung cake and crop waste. The uncertainties on fuelwood, dung cake, and crop waste user population fractions for rural and urban regions were derived as differences in the national user population fractions reported from *NFHS* [2001] and Census of India (2001; see http://www.censusindia.net/2001housing/S00-018.html). These were 12, 34, and 38%, respectively, for fuelwood, dung cake, and crop waste rural user population fractions, and were applied on each state; corresponding values for urban regions were 2, 30, and 100%, applied on statewise urban user population fractions.

[13] The thermal efficiency for various stove-fuel systems and calorific values of different fuels (Table 2) [*Ahuja et al.*, 1987; *Mukunda et al.*, 1988; *Joshi et al.*, 1989; *Kandpal and Maheshwari*, 1995; *Kandpal et al.*, 1995; *Koopmans and Koppejan*, 1997; *Ravindranath and Ramakrishna*, 1997; *Gupta et al.*, 1998; *Venkataraman and Rao*, 2001] were used to estimate the energy consumption (PJ yr^{-1}) and fuel demand in mass units (Tg yr^{-1}).

4. Measurement of Sulfur Dioxide Emission Factors From Biofuel Combustion

[14] The traditional one-pot stove extensively used by rural households in India, from a review of stove use [*Ministry of Non-Conventional Energy Sources*, 2001], was adopted for the biofuel combustion experiments. Surveys of crop waste used as biofuel indicate a predominance of waste with higher energy density, like stalks of oilseeds, fiber crops, and pulses [*Ravindranath and Hall*, 1995]. Fuels selected included two species of wood, dung cake, and 10 types of widely used crop waste (Table 3). All fuels were cut to standard sizes and sun-dried for 7 days, and representative random samples were made.

[15] SO_2 was measured with the U.S. EPA standard spectroscopic method [e.g., *Luke*, 1997], using a dilution sampler (Figure 2), optimized for quantitative emission factor measurement [*Venkataraman and Rao*, 2001; *Venkataraman et al.*, 2004]. In the dilution sampler, combustion gases from the stove are entrained into a duct, following dilution provided by an induced draft fan, and introduced into gas samplers. To avoid the loss of

Table 3. Traditional Single Pot Mud Stove Fuel System Used in Study

Biofuel Categories	Types	Source
Fuelwood	acacia nilotica, Eucalyptus	local market and IIT campus
Dung cake	cow dung patties	Eksaal village, Maharashtra
Crop waste	straw of rice and wheat stalks of soyabean and mustard	Eksaal village, Maharashtra, Rajim village, Chattisgarh
	stalks of tur (pigeon pea) and cotton, stems and roots of tobacco and Sugarcane	Pandhari and Kapsi villages, Maharashtra
	stalks of Jute	Bankura village, West Bengal
	straws of Ragi (finger millet)	Davanagrera village, Karnataka

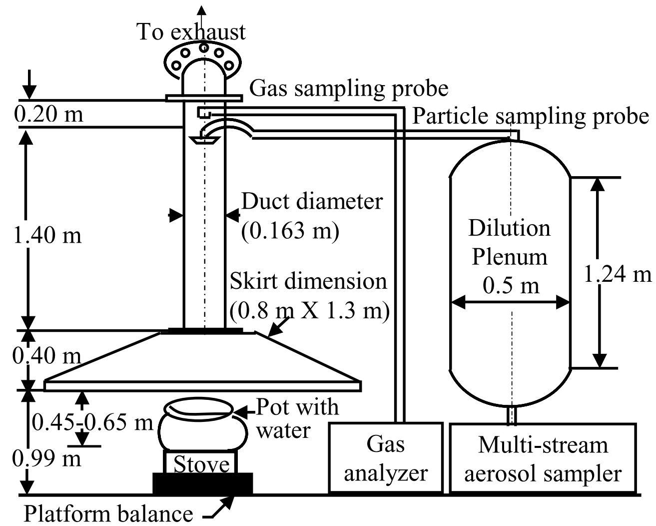

Figure 2. Schematic diagram of the experimental set-up designed for quantitative emission factor measurement.

SO_2, the Teflon tube was used to draw the gas from the duct to the inlet of the gas sampler. The total gas flow rate in the duct was calculated from the midpoint velocity, measured each minute by a platinum hot-film sensor, calibrated over a velocity range of 0.5–3.5 m s^{-1}, using a Pitot tube in a converging wind tunnel. The pollutant emitted over the burn experiment (grams) was calculated from the STP-corrected gas flow rate (m^3 h^{-1}) and the pollutant concentration (g m^{-3}), and time of burn (hours), and divided by fuel combusted (kilograms) to calculate the emission factor (grams of SO_2 (kilograms of fuel burned)$^{-1}$).

[16] The system was optimized to minimize combustion modification from the induced draft by ensuring that combustion temperature was not reduced by dilution from the

Table 4. Thermal Parameters and Emission Factors of Sulfur Dioxide From Biofuel Combustion

Biofuel Types	Fuel, kg	Time, min	Dilution Ratio (DR)[a]	Burn Rate ($\check{R}$), kg h^{-1}	Combustion Temperature, °C	Thermal Efficiency, %	$(Air/Fuel)_{actual}$[b]; Stoichiometric Ratio[c]	SO_2 Emission Factors 5,[d] g kg^{-1}
				Fuelwood				
Acacia[e]	0.30	15	16	0.9	525	13	3.0; 0.7	0.08
Eucalyptus[e]	0.25	15	24	0.9	406	14	2.0; 0.3	0.03
Dung cake[f]	0.30	15	40	1.3	479	11	1.6; 0.4	0.88
				Crop Waste				
Soyabean stalk[f]	0.46	15	18	1.6	383	10	5.6; 1.1	0.30
Mustard stalk[f]	0.43	15	14	1.5	430	13	4.8; 0.9	0.20
Tobacco stem[e]	0.30	15	21	0.9	449	15	2.4; 0.5	0.20
Rice straw[f]	0.43	15	21	1.9	368	11	2.8; 0.7	0.15
Ragi straw[f]	0.45	15	19	1.8	358	12	3.1; 0.5	0.13
Wheat straw[f]	0.46	15	21	1.9	395	11	2.3; 0.4	0.10
Cotton stalk[f]	0.48	15	21	1.5	495	18	5.0; 0.9	0.08
Jute stalk[e]	0.60	15	13	1.1	424	20	2.8; 0.4	0.05
Tur stalk[f]	0.47	15	18	1.5	423	16	4.3; 0.7	0.05
Sugarcane[e] root and stem	0.30	15	21	0.9	501	19	2.1; 0.4	0.05
Ave ± SD[g]	**0.44 ± 0.09**	**15**	**18 ± 3**	**1.5 ± 0.4**	**422 ± 49**	**15 ± 4**	**3.5 ± 1.3; 0.6 ± 0.2**	**0.13 ± 0.08**

[a]Dilution ratio calculated based on combustion temperature, corrected for radiation losses, and duct temperature; DR = $(T_{comb} - T_{duct})/(T_{duct} - T_{amb})$, where T_{comb} is combustion temperature, T_{duct} is duct temperature, and T_{amb} is ambient temperature in degrees Kelvin (K).

[b]$(Airflow\ rate/fuel\ flow\ rate)_{actual}$, where airflow rate through the stove was calculated as $\{[(Q\ m^3\ h^{-1} \times 298\ K \times \rho\ kg\ m^{-3})/((DR + 1) \times T_{duct}\ K)] - \check{R}\ kg\ h^{-1}\}$, where Q is the gas flow rate through duct in m^3 h^{-1}, ρ is air density, i.e., 1.25 kg m^{-3} at 298 K, and $\check{R}$ is the fuel flow rate in kg h^{-1}.

[c]Calculated as $[(air/fuel)_{actual}/(air/fuel)_{stoichiometric}]$, where $(air/fuel)_{stoichiometric}$ range is 4.4–5.9 for wood and 4.0–7.0 for crop wastes, and is 4.0 for dung cake based on volatiles empirical formula derived as $CH_{1.5}O_{0.8}$ for wood, $CH_{1.9}O_{0.8}$ for crop wastes, and $CH_{2.2}O_{1.2}$ for dung cake from fuel composition, assuming evolved carbon equals [total carbon $_{ultimate\ analysis}$ − fixed carbon $_{proximate\ analysis}$].

[d]Emission factors from single experiment for each biofuel type with measured nominal uncertainty of 5%, determined separately as 1 standard deviation from the mean of three experiments for dung cake.

[e]Extraction flow rate 0.02 m^3 s^{-1}.

[f]Extraction flow rate 0.05 m^3 s^{-1}.

Table 5. Comparison of Present Estimates of Biofuel Consumption for Cooking (Tg yr^{-1}) With Literature

	Present Study	*Streets and Waldhoff* [1998][a]	*Reddy and Venkataraman* [2002][a]	*Streets et al.* [2003][a]	*Yevich and Logan* [2003][a]	*Bond et al.* [2004][a]	*Smith et al.* [2000][a]
Base year	2000–01	1990	1996–97	2000	1985	1996	1990
Total	379 (247–584)[b]	521	538	421	399	478	286
	(54%)[c]	[573][d]	[581]	(100%)	[518]	[516]	[342]
					(95%)	(100%)	
Biofuel Types							
Fuelwood	281 (192–409)	271	302	316[e]	220	265	169
	(46%)	[298]	[326]	(100%)	[286]	[286]	[202]
Dung-cake	62 (35–108)	124	121	105	93	128	54
	(74%)	[136]	[131]	(200%)	[120]	[138]	[65]
Crop waste	36 (20–67)	126	115		86	85	63
	(86%)	[139]	[124]		[112]	[92]	[75]

[a]Including biofuel used for cooking and water heating.
[b]Central value and uncertainty ranges in parentheses were calculated at 95% CI.
[c]Values are the 95% CI as percentage of central value.
[d]Values are upgraded biofuel consumption estimates for the base year 2000–2001, using the ratio of rural population for current base year and the reported base of the study. These ratios were 1.3, 1.2, and 1.1 for base years 1985, 1990, and 1996, respectively.
[e]Including both the fuelwood and crop waste.

draft. The draft flow rate and resulting stoichiometric ratio ($(air/fuel)_{actual}/(air/fuel)_{stoic}$)were chosen such that the combustion temperature was maintained at its maximum value. The actual air-fuel ratio was calculated using the air flow rate through the stove, calculated as the difference between the gas and fuel flow rates (kg h^{-1}) through the stove. The gas flow rate through the stove was obtained by correcting the measured gas flow rate through the duct, using the measured dilution ratio. The radiation-corrected combustion temperature [*Rohsenow and Hunsaker*, 1947] and midpoint duct temperature were used to calculate dilution ratios, which varied from 15 to 40 (Table 4). The stoichiometric air-fuel ratio was derived from measured fuel composition from proximate and ultimate analysis of fuels used in this study.

[17] It was found that dilution from external air reduces combustion temperature. Thus an extraction draft rate was chosen, corresponding to a duct midpoint velocity of 3 m s^{-1} for dung cake, rice straw, and mustard stalks and 1.24 m s^{-1} for wood and jute stalk, at which combustion temperature was maximum. Reduction in combustion temperature occurred around stoichiometric ratios of 0.3–0.9 (Table 4), where air availability was far lower than the stoichiometric requirement, indicating poor mixing in the combustion zone.

[18] The burn cycle adapted from recent variants of the water boiling test [*Volunteers in Technical Assistance*, 1985; *Smith et al.*, 2000; *Venkataraman and Rao*, 2001] consisted of heating 0.5 kg water from room temperature to the boiling point and simmering for 5 min, leading to a total burn time of 15 min. The burn rates used were derived from literature, i.e.,1.0 kg h^{-1} for wood, 1.3 kg h^{-1} for dung, 1.6 kg h^{-1} for stalks such as mustard, and 1.8 kg h^{-1} for straws such as rice and wheat that are typical in rural cooking practice [*Smith et al.*, 1983, 2000]. The burn cycle was designed to simulate actual cooking practice, with multiple charges two for wood and dung cake at 5-min intervals and four for crop wastes at 3-min intervals. The burn cycle includes both the high and low power phases.

[19] A sampling rate of 0.5 lit min^{-1} was used in the pulsed fluorescence spectrometer for SO_2 (Model 8850, Monitor Labs, U.S.A.). The pulsed fluorescence spectrometer was obtained on loan from the collaborators at the University of Maryland. Potassium carbonate impregnated filters were used to set the zero of the instrument before single-point calibration prior to each experiment. The multipoint calibration was carried out at Maryland before and after the entire set of experiments, and the calibration factor was multiplied into the single-point calibration done at IIT Bombay, prior to each experiment, using a span SO_2 gas of 9.2 ppm. The span gas was added at the inlet of the instrument using a small (10 cm) tube to avoid the loss of SO_2 during calibration. The lower detection limit was 0.05 ppm and relative precision, as 1 standard deviation from the mean of three repeat experiments, was 5%.

5. Propagation of Uncertainties

[20] A specific goal of the methodology developed, for estimating biofuel consumption and associated emissions, was that the uncertainties in all input variables were characterized and propagated to obtain upper and lower bounds (at 95% confidence). For multiplicative independent variables, used to calculate biofuel consumption, the relative precision was propagated in quadrature to obtain the uncertainty. For national level estimates, the absolute precision in statewise variables was linearly added to obtain the uncertainty, as the individual state values were derived from common input data and therefore were not independent. The 95% confidence intervals for biofuel use were calculated as 1.96 times the absolute precision. The lower/upper bounds were derived assuming lognormally distributed uncertainties, following *Streets et al.* [2003].

6. Results and Discussion

6.1. Cooking Energy and Biofuel Consumption in India

[21] Cooking energy consumption was estimated separately for rural and urban regions. The total cooking energy consumption for India for 2000 was 6325 PJ with rural population using about 84%. This reflects both the large rural population and the use of low-efficiency biofuel cooking devices. Also, cooking energy consumption from

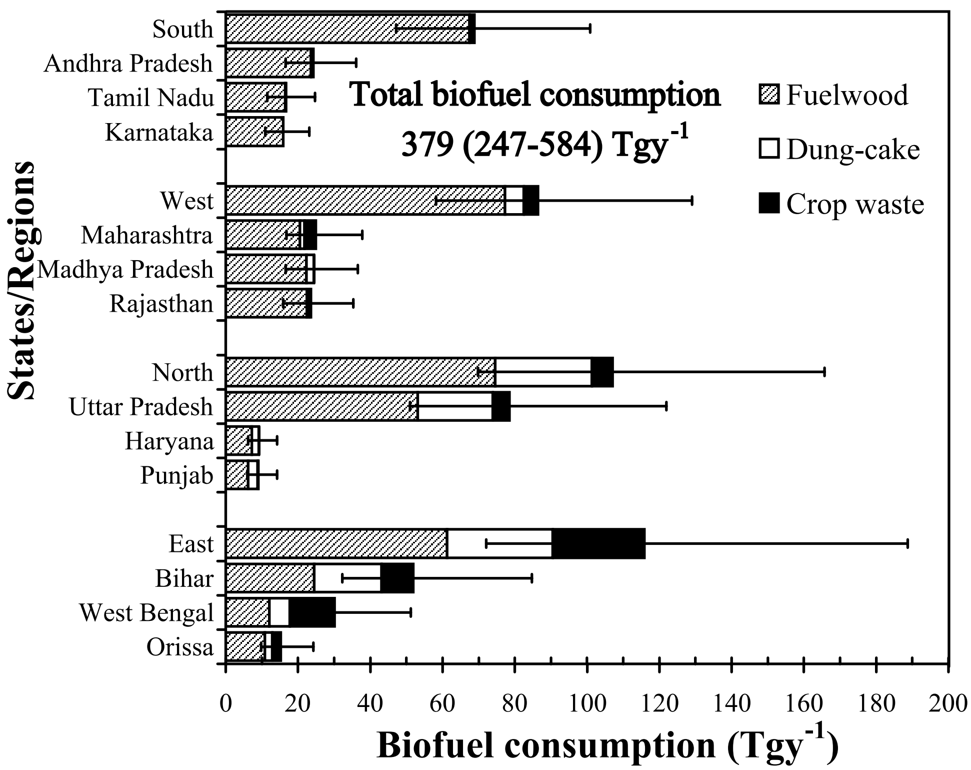

Figure 3. Biofuel consumption for cooking in major states and regions of India showed high fuelwood consumption in all regions, and large dung cake and crop waste consumption in northern and eastern states; error bars were estimated at 95% CI.

biofuels in India was predominant, 92% (5825 PJ), with a minor contribution of 8% (500 PJ) from fossil fuels.

[22] Total biofuel consumption was estimated as 379 (247–584) Tg yr^{-1}, resulting in a biofuel mix of 74:16:10% of fuelwood, dung cake, and crop waste, respectively (Table 5). National central value and uncertainty range of fuelwood consumption were 281 (192–409) Tg yr^{-1}, with predominance in all regions (Figure 3). The central values and uncertainty ranges of dung cake and crop waste consumption in India were 62 (35–108) Tg yr^{-1} and 36 (20–67) Tg yr^{-1}, with northern and eastern states (mainly Uttar Pradesh, Bihar, and West Bengal) contributing about 70%. The high biofuel consumption in northern and eastern regions reflects higher per capita food consumption and significant use of dung cake and crop waste (as described in section 3.0) compared to the western and southern regions.

[23] Our biofuel use estimate for cooking was lower than previous estimates (compared in Table 5), using per capita biofuel consumption reported together for cooking and water heating from REDB related energy use surveys [*Joshi et al.*, 1992; *Sinha et al.*, 1998], and assumed various user populations [*Streets and Waldhoff*, 1998; *Reddy and Venkataraman*, 2002; *Streets et al.*, 2003; *Bond et al.*, 2004]. However, it compares well with the study by *Smith et al.* [2000], which uses per capita biofuel consumption from a different compilation of surveys [*Integrated Rural Energy Planning Programme* (*IREP*), 1992]. The IREP per capita biofuel consumption values were significantly lower (about 0.4–0.5 times) than those from REDB, while both surveys were similar in terms of sample size (about 650 villages), agroclimatic zones (14 zones) considered, and base year (1985). This is reflective of the large uncertainty inherent in the energy survey approach.

[24] As the new methodology developed here, based on food consumption statistics and energy required for food preparation, is entirely different from the energy-survey based methodology used so far, it is pertinent to compare the uncertainties in the current estimate with those in

Table 6. Comparison of Measured Emission Factors (g kg^{-1}) for Biofuel Combustion With Literature

Biofuel Types	Present Study	*Streets et al.* [2003]	*Reddy and Venkataraman* [2002]	*Olivier et al.* [2001] (EDGAR 3.2)	*Garg et al.* [2001]	*Zhang et al.* [2000]	*Streets and Waldhoff* [1998]
Fuelwood	0.06 (0.03–0.08)[a]	0.18–1.11	0.48	0.20[b]	0.80	0.01	0.60
Dung cake	0.88	0.18–1.11	0.84	4.70[b]	0.60		6.00
Crop waste	0.13 (0.05–0.20)[a]	0.18–1.11	0.48	0.90[b]	0.60	0.22	

[a]Central values and ranges for different biofuel species from Table 4.
[b]Values reported in g GJ^{-1} and converted to g kg^{-1} using suitable calorific values for respective biofuels, 15,100, 15,500 and 11,760 KJ kg^{-1} for wood, crop waste, and dung cake, respectively.

Table 7. Central Value and Uncertainty Ranges of Biofuel Consumption for Cooking in India, Emission Factors of Black Carbon and Sulfur Dioxide

		Emission Factors, $g\ kg^{-1}$	
Biofuel Types	Biofuel Burned, $Tg\ yr^{-1}$	BC	SO_2
Total biofuel	379 (247–584)[a] (54%)[c]	0.59 (0.26–1.31)[b] (122%)	
Fuelwood	281 (192–409) (46%)		0.06 (0.05–0.06)[d] (10%)
Dung cake	62 (35–108) (74%)		0.88 (0.80–0.97)[d] (10%)
Crop waste	36 (20–67) (86%)		0.13 (0.06–0.25)[d] (97%)

[a]Central value and uncertainty ranges in parentheses are calculated at 95% CI.
[b]*Andreae and Merlet* [2001].
[c]Values are the 95% CI as percentage of central value.
[d]Measured emission factors are from Table 4.

previous estimates. The lognormal distribution of uncertainties was (following *Streets et al.* [2003]) resulted in an asymmetric 95% CI around the mean, if examined on a linear scale. We therefore express the uncertainties as a factor operated on the mean. The national level uncertainty in biofuel use (about 50%) was significantly lower than the uncertainties (100–200%) in previous work [*Streets et al.*, 2003; *Yevich and Logan*, 2003; *Bond et al.*, 2004].

[25] The most significant finding was the biofuel mix of 74:16:10% of $Tg\ yr^{-1}$ fuelwood, dung cake, and crop waste use, in contrast with that of 56:23:21% of $Tg\ yr^{-1}$ in previous studies [*Sinha et al.*, 1998; *Reddy and Venkataraman*, 2002; *Smith et al.*, 2000]. From the new National Family Health Survey [*NFHS*, 2001], in which respondents were queried on the predominant fuel they used for cooking, 73% of the rural population used fuelwood, while only 8% each used dung cake and crop waste and the remaining 11% used other fuels (e.g., coal LPG and kerosene). This is in sharp contrast to most previous studies, in which 100% of the rural population was assumed to use all three biofuels.

6.2. Emission Factors of Sulfur Dioxide From Biofuel Combustion

[26] Sulfur dioxide emission factors ranged over 0.03–0.08 $g\ kg^{-1}$ for wood fuels, 0.88 $g\ kg^{-1}$ for dung cake combustion, and 0.05–0.20 $g\ kg^{-1}$ for crop waste (Table 4). These SO_2 emission factors, measured for the first time with pulsed fluorescence analyzer, are higher for wood and lower for crop wastes than those measured by *Zhang et al.* [2000] using the wet chemical method (Table 6). Fuel sulfur content is expected to range from 0.01 to 0.04% for wood, 0.01 to 0.05% for crop waste, and 0.07 to 0.10% for dung cake [*Smith et al.*, 2000; *Venkataraman and Rao*, 2001; *Reddy and Venkataraman*, 2002]. This implies that sulfur emitted as SO_2 ranged from 6 to 50% of fuel sulfur content.

[27] It is not appropriate to quantitatively compare the present measurements with measurements in the literature, because of the multiple measurement methods involved. Previous measurements of SO_2 emission factors have been made using an electrochemical sensor [*Ballard-Tremeer and Jawurek*, 1996] and diffusion dosimeter colorimetric tubes [*Smith*, 1988]. These studies chose methods that were inexpensive and operationally easy in remote areas, but with relatively large uncertainty [*Smith*, 1987]. These emission factors were adopted widely in current inventories [*Streets and Waldhoff*, 1998; *Olivier et al.*, 2001]. The other approach used in inventories was to derive the emission factor from fuel sulfur content and an assumed fraction of sulfur remaining in ash, rather than direct measurement [*Garg et al.*, 2001; *Reddy and Venkataraman*, 2002]. Discrepancies have arisen especially for emissions from dung cake [*Streets and Waldhoff*, 1998; *Olivier et al.*, 2001] for which SO_2 emission factors significantly exceeded the typ-

Table 8. Comparison of Black Carbon Emissions From Biofuels From India With Literature

	Black Carbon Emissions, $Gg\ yr^{-1}$				
	Present Study	*Dickerson et al.* [2002]	*Reddy and Venkataraman* [2002]	*Bond et al.* [2004]	*Streets et al.* [2003]
Base year	2000–2001	2000–2001	1999	1996	2000
			Biofuel Types		
Biofuel	220 (65–760)[a] (245%)[b]	420	207 [215][c]	330 [340][c]	421 (486%)
Fuelwood	165 (50–530) (220%)		123 [130]	177 [180]	316[d]
Dung cake	35 (10–140) (300%)		30 [30]	68 [70]	105
Crop waste	20 (5–90) (350%)		54 [55]	85 [90]	

[a]Central value and uncertainty ranges in parentheses are calculated at 95% CI.
[b]Values are the 95% CI as percentage of central value.
[c]Values are upgraded for the current base year using the rural population.
[d]Value includes the emissions from fuelwood and crop waste combustion.

Table 9. Comparison of Sulfur Dioxide Emissions From Biofuel Combustion From India With Literature

	Sulfur Dioxide Emissions, Gg yr^{-1}				
	Present Study	*Streets and Waldhoff* [1998]	*Garg et al.* [2001]	*Reddy and Venkataraman* [2002]	*Streets et al.* [2003]
Base year	2000–2001	1990	1995	1996–1997	1995
			Biofuel Types		
Biofuels	75 (36–160)[a] (113%)[b]	880 [1038][c]	278 [300]	300 [312]	229 (199%)
Fuelwood	15 (10–30) (100%)			145 [150]	123[d]
Dung cake	55 (25–110) (100%)			101 [105]	106
Crop waste	5 (1–20) (300%)			55 [57]	

[a]Central value and uncertainty ranges in parentheses are at 95% CI.
[b]Values are the 95% CI as a percentage of central value.
[c]Values were upgraded for base year 2000–2001 by using rural population.
[d]Values include the emissions from fuelwood and crop waste combustion.

ical sulfur content of dung (0.07–0.10%) [*Smith et al.*, 2000; *Venkataraman and Rao*, 2001; *Reddy and Venkataraman*, 2002]. The present work represents an effort to bridge this gap, and provide direct measurements of SO_2 emission factors from biofuel combustion using a standard method.

6.3. Emissions of Black Carbon and Sulfur Dioxide From Biofuel Combustion

[28] BC emissions from biofuel combustion in India were estimated by combining the biofuel use estimates developed here with the respective emission factors. Emission factors for BC from biofuels reported by *Andreae and Merlet* [2001] is 0.59 ± 0.37 g kg^{-1} for all three types of biofuel (Table 7). BC emission factors for biofuels differed in various studies, and we chose to use the one compiled by *Andreae and Merlet* [2001]. A more recent study by *Sheesley et al.* [2003] measured BC emission factors of 0.05–0.14 g kg^{-1} (average 0.09 g kg^{-1}) from dung cake, rice straw, and jackfruit branches and 0.35 g kg^{-1} from coconut leaves. These are lower than previous measurements [*Cachier et al.*, 1996], possibly from the relatively lower combustion temperatures and higher burn rates used in these experiments than those typical in biofuel cooking [*Smith et al.*, 1983; *Ramachandra et al.*, 2000], and are therefore not included in the emission factor range used here. Uncertainty was estimated as 95% CI bounds on emissions. The central value lower and upper bounds on the emissions were derived using the assumption of a lognormal distribution of uncertainties in activity data and emission factors, following *Streets et al.* [2003].

[29] Total BC emissions from biofuels from India were estimated as 220 (65–760) Gg yr^{-1} (Table 8), as central value and uncertainty range. Fuelwood accounts for 165 (55–530) (75%), dung cake for 35 (10–140) Gg yr^{-1} (16%), and crop waste for 20 (5–90) Gg yr^{-1} (9%) of BC emissions from biofuel combustion from India. Our BC emissions from biofuel combustion are lower than previous studies [*Dickerson et al.*, 2002; *Streets et al.*, 2003; *Bond et al.*, 2004], for the year 2000 (Table 8). Though the uncertainty in our biofuel use estimate is low (54%), the large uncertainty in the associated BC emission factor (122%) (Table 7) resulted in an uncertainty of 245% in BC emissions from biofuel combustion, implying the need for comprehensive measurement of emission factors from a set of biofuels widely used in India.

[30] SO_2 emissions were estimated for biofuel combustion in India by combining activity data of biofuels with respective measured emission factors. The estimated SO_2 emissions from biofuel combustion were 75 (36–160) Gg yr^{-1} as the central value and uncertainty ranges (Table 9). The major contribution was from dung cake, 55 (25–110) Gg yr^{-1} (73%), followed by fuelwood, 15 (10–30) Gg yr^{-1} (21%), and crop waste, 5 (1–20) Gg yr^{-1} (6%). The central value of SO_2 emissions is about a factor of 3 lower than that in other recent studies (Table 9), because of the major reduction in SO_2 emission factors (about factor of 10) measured by standard method using pulsed fluorescence spectroscopy, compared to those previously used [*Streets and Waldhoff*, 1998; *Garg et al.*, 2001; *Reddy and Venkataraman*, 2002]. Large uncertainties in total SO_2 emissions (113%) were from the uncertainty in both the activity data and emission factors for crop waste combustion (Table 6), which ranged widely for different crop types.

7. Conclusions

[31] In this study, a new methodology was developed for estimating biofuel use for cooking, based on food consumption statistics and specific energy requirements for food preparation. Total biofuel consumption in India for base year 2000 was estimated as 379 (247–584) Tg yr^{-1}. A significant result is the large fuelwood use, with a national average biofuel mix of 74:16:10% of Tg yr^{-1} for fuelwood, dung cake, and crop waste, respectively. Importantly, the national level uncertainty in biofuel use, resulting from error propagation, was bounded at about 50%. BC emissions from biofuel combustion from India were estimated at 220 (65–760) Gg yr^{-1}, dominated by fuelwood combustion (75%), with dung cake and crop waste accounting for 16% and 9%, respectively. The large uncertainty in BC emission factors from biofuel combustion now governs the emissions estimate and implies the need for comprehensive measurements of emission factors from widely used biofuels, based on actual rural cooking practice. SO_2 emission factors, measured for the first time using pulsed fluores-

cence spectrometry, ranged from 0.03 to 0.08 g kg^{-1} for woods and from 0.05 to 0.20 g kg^{-1} for crop waste straws and woody stalks, and was 0.88 g kg^{-1} for dung cake combustion. SO_2 emissions from biofuel combustion from India were 75 (36–160) Gg yr^{-1}, had an uncertainty of 113%, and were about a factor of 3 lower than that estimated in recent studies. The biofuel use estimates developed in this work must be incorporated into regional and global emissions inventories for a comprehensive list of gaseous and particulate pollutants, to assist uncertainty reduction in regional and global climate assessment.

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R. Banerjee, Energy Systems Engineering, Indian Institute of Technology Bombay, Powai, Mumbai- 400 076, India.

R. R. Dickerson and J. W. Stehr, Department of Meteorology, University of Maryland, College Park, MD 20742, USA.

G. Habib and C. Venkataraman, Department of Chemical Engineering, Indian Institute of Technology Bombay, Powai, Mumbai-400 076, India. (gazala@cc.lltb.ac.in)

M. Shrivastava, Center for Environmental Science and Engineering, Indian Institute of Technology Bombay, Powai, Mumbai 400 076, India.