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32. Supported by the National Institute of Dental and Craniofacial Research–NIH grants to J.M.-O.: R01-DE-13414 and R21-DE-15332. We thank C. Shuler, M. Snead, and C. Wuenschell from the Center for Craniofacial Molecular Biology, University of Southern California for reading this manuscript before its submission. We thank J. Aizenberg from the Bell Labs, Lucent Technologies, New Jersey, for useful discussions.

Residential Biofuels in South Asia: Carbonaceous Aerosol Emissions and Climate Impacts

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High concentrations of pollution particles, including "soot" or black carbon, exist over the Indian Ocean, but their sources and geographical origins are not well understood. We measured emissions from the combustion of biofuels, used widely in south Asia for cooking, and found that large amounts of carbonaceous aerosols are emitted per kilogram of fuel burnt. We calculate that biofuel combustion is the largest source of black carbon emissions in India, and we suggest that its control is central to climate change mitigation in the south Asian region.

The effect of pollution particles measured over the Indian Ocean (*1–3*) on the regional atmospheric radiation balance is about 10 times the effect of greenhouse gases (*4*), leading to a large cooling at Earth's surface and a strong heating of the atmosphere. Large changes in atmospheric absorption and the radiation balance could affect rainfall patterns (*5*), which in turn could contribute to increased intensity of droughts and floods, for example, in China and potentially in neighboring India. Emissions control of light-absorbing soot, in addition to control of greenhouse gases, has been suggested as a measure to slow global warming (*6*, *7*). Soot or black carbon (BC) emissions in the south Asian region arise from fuel combustion for transportation, industrial, and residential uses. Here we find, from carbonaceous aerosol measurements and fuel use estimates, that the combustion of solid biofuels—such as wood, agricultural waste, and dried animal manure in cooking stoves—is the largest source of BC emissions in India. We therefore suggest that the control of these emissions through cleaner cooking technologies, in addition to reducing health risks to several hundred million users, could be of crucial importance to climate change mitigation in south Asia. Similar considerations may also apply to other regions of Asia, as well as Africa and South America, where residential biofuel combustion is prevalent.

Whereas greenhouse gases trap heat in Earth's atmosphere, aerosols (particles with diameters from 0.01 to 20 µm) can cool or heat the atmosphere, depending on their light-scattering or light-absorbing nature. Atmospheric aerosols contain acidic species (including sulfates and nitrates), carbonaceous constituents [BC and organic carbon (OC)], inorganic matter (fly ash), sea salt, and dust. Light-absorbing sp^2-bonded carbon, measured by change in light transmittance or reflection, is termed BC; refractory graphitic carbon, measured by thermal evolution under high-temperature oxidation (sometimes with simultaneous change in light transmittance or reflection), is termed elemental carbon (EC) (*8*, *9*). Most measurements of EC are treated as equal to BC.

High atmospheric concentrations of BC were recently measured in the south Asian region (*1–3*). These aerosols were attributed largely to fossil fuel emissions [according to the chemical mass balance principle (*10*)], but the particulate chemical signatures typical of fossil fuel and biomass burning sources (*1*, *2*) were not specific to south Asia. At the same time, estimates of energy use (*8*, *11–13*) showed a large amount of biofuel combustion for cooking, not believed so far to be a significant source of climate-forcing pollutants in terms of its negligible contribution to global CO_2 emissions (*14*). BC emission factors, not yet published for biofuel cooking stoves, are typically estimated from reported particle emission factors in inventory and review studies (*8*, *11–13*, *15*). The quantity and composition of particulate emissions (*16*) and their radiation absorption (*17*) depend on fuel species, moisture content and size, burn rate, and firing practice, making them highly source-specific. We report measurements of carbonaceous aerosol emission factors (in terms of grams of carbon per kilogram of fuel burnt) from combustion of a variety of biofuels widely used in the south Asian region, estimate their contribution to regional and global BC emissions, and discuss climate implications.

We conducted laboratory combustion experiments with four species of wood, six types of crop waste, and dried cattle manure (table S1), widely used as biofuels in India (*13*), in a traditional one-pot stove estimated to account for more than 80% of Indian stove use (*13*). A dilution sampler, developed and optimized in previous work (*18*, *19*), was used to achieve postcombustion quenching and gas/particle partitioning that would occur in actual indoor environments (fig. S1A). Emissions were entrained into a hood, and particles smaller than 2.5 µm aerodynamic diameter (PM-2.5), sampled through a probe, were collected on prebaked quartz-fiber filters by a cyclone-inlet particle sampler during combustion experiments (fig. S1B) and were analyzed for BC and OC to calculate emission factors (*18*).

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The burn cycle, designed to simulate local cooking practice (13, 19, 20), included low- and high-power phases and involved heating 0.5 kg of water from room temperature to its boiling point, then simmering for 5 min. To capture variation in rural cooking practice (13, 20), we burned wood at average burn rates of 0.9 to 2.0 kg hour^{-1} (using two fuel charges), dried cattle manure at 1.3 kg hour^{-1} (using two charges), and crop waste at 1.1 to 1.9 kg hour^{-1} (using three charges) (table S1), with fuel charges suited to fit the stove combustion chamber and deliver the required power. We also burned two fossil fuels—liquefied petroleum gas (LPG) and kerosene—at 0.1 and 0.2 kg hour^{-1}, respectively, in stove burners used widely for residential cooking. Measured combustion temperatures, corrected for the heat lost by radiation [following (19)], varied from 300° to 670°C for different fuels and combustion conditions. Ambient air diluted the combustion gases at dilution mass ratios of 10 to 40 during combustion of different biofuels (table S2). The estimated equivalence ratios (table S1) were greater than 1, implying highly fuel-rich flame conditions—that is, the actual fuel-to-air ratios in the flame were higher than the corresponding stoichiometric ratios for complete oxidation of fuel constituents (table S2).

Measured BC emission factors ranged from 0.38 to 0.62 g kg^{-1} from wood combustion [for varying burn rates (0.9 to 2.0 kg hour^{-1}) and corresponding combustion temperatures (396° to 644°C)] and from 0.12 to 0.17 g kg^{-1} from straw and dried cattle manure; these values were at the lower end of the range 0.3 to 1.4 g kg^{-1} estimated in recent inventories (8, 12, 13, 15). Measured OC emission factors were 0.17 to 4.69 g kg^{-1}, again lower than estimates of 1.7 to 7.8 g kg^{-1} in inventory studies (8, 12, 13, 15), and varied with fuel type, increasing systematically with burn rate (table S1). On the basis of fuel burnt, BC and OC emission factors from kerosene and LPG stoves were lower than those from biofuel stoves by a factor

of 3 to 50 (table S1)—or, in terms of megajoules of heat recovery, lower by a factor of 15 to 150—because of the higher thermal efficiencies of kerosene and LPG stoves.

In thermal-optical measurement methods (18), such as the one used in this study, the relative amount of evolved carbon allocated as BC and OC is sensitive to the temperature program chosen, particularly for wood smoke samples (9). The method adopted here uses a maximum temperature of 870°C (fig. S2), as opposed to 550° to 750°C, for pyrolysis under pure helium to correct for the error in BC from charring of OC, and results in lower BC values than the low-temperature methods would have yielded (9). Artifact OC enhancement was measured through OC adsorption on a quartz-fiber filter in a parallel stream, after particle filtering (18), and subtracted from the sample OC (fig. S3 and table S3). Artifact OC was specifically high for two wood species, jamun and neem (table S3). An uncertainty in this measurement would not change the BC but would increase the OC emission factors for these species. This would reduce the spread in the BC/OC ratio from different wood species but would not change the mean value significantly, nor would it alter our overall conclusions.

BC constituted 5 to 52% of PM-2.5 mass, consistent with the observation that large BC fractions are possible in emissions from small cooking fires (8). This range is also substantially larger than the 3 to 6% reported from large fires in different forest and grassland classes (15, 21). OC fractions of 14 to 52% from biofuel were correspondingly lower than the 60 to 70% reported from open biomass burning (15). We found that BC and OC constituted 51 to 67% of biofuel PM-2.5; together with potassium and levoglucosan, they are reported to constitute 70 to 85% of PM-2.5 from open burning. Our data yielded BC/OC ratios of 0.11 to 3.53 in biofuel aerosols; these values are significantly higher than the ratios of 0.06 to 0.14 reported from open burning and are indistinguishable from

the ratio of 1.0 reported from diesel transport and household coal emissions (2).

Although the formation of BC and OC in biomass and biofuel fires is not well understood and is beyond the scope of this work, it would depend on variables such as burn rate, air-fuel ratio, flame turbulence, and combustion temperature. We estimate biomass burn rates in forest fires to range from 100 to 3000 kg hour^{-1} on the basis of reported experimental burns (22). Typical burn rates in fireplaces and space-heating stoves (23, 24) are 2 to 9 kg hour^{-1}, significantly higher than in the biofuel cooking fires studied here. The low burn rates in biofuel cooking fires result in very low air induction into the flame by thermal buoyancy, as evidenced by the large equivalence ratios (table S1) characteristic of fuel-rich flame conditions, which can result in high BC and low OC formation. We therefore emphasize that small biofuel fires are distinct in nature from large fires and that a more complete understanding of their aerosol emissions, especially chemical and optical characteristics relevant to climate, remains to be gained.

The large BC content in emissions from biofuel combustion has the potential for significant atmospheric impact given the extensive use of these fuels in India (13). In Table 1, BC emissions from biofuel combustion in India are estimated from the emission factors we measured, together with fuel usage data for India (13) and other world regions (8, 25). Because biofuel combustion for cooking occurs most often in indoor environments, we consider in these estimates the fraction of emissions that penetrate to the ambient air, estimated at about 80% on the basis of typical ventilation and particle deposition rates in rural kitchens (26). BC emissions from biofuel combustion in India are 172 to 340 Gg year^{-1}, hence the relative contributions to total BC emissions from fossil fuel, open burning, and biofuel combustion are 25%, 33%, and 42%, respectively. The corresponding OC emissions are 582 to 1683 Gg year^{-1}, so that

Table 1. Black carbon (BC) emissions from biofuel combustion in India, Asia, and the world. Estimates of BC emissions from biofuel combustion made use of emission factors (g kg^{-1}) from table S1. In the rightmost column, the total includes BC emissions from forest, savanna, and crop waste open burning as well as fossil fuel combustion.

Region	Base year	Biofuel consumption (Tg year^{-1})			Black carbon emissions (Gg year^{-1})				BC source ratio (biofuel/total) (%)
		Fuelwood	Dried cattle manure	Crop waste	Fuelwood	Dried cattle manure	Crop waste	Total biofuel	
India	1995	281* (192–409)‡ (13)	62* (35–108)‡ (13)	36* (20–67)‡ (13)	143* (75–272)‡	8* (3–17)‡	21* (9–51)‡	172* (87–340)‡	44†
Asia		800–930§ (8, 25)	130–200§ (8, 25)	430–545§ (8, 25)	400–470	15–25	220–280	635–775	30‡
Global		1324–1615§ (8, 25)	150–410§ (8, 25)	442–707§ (8, 25)	670–820	20–50	230–360	920–1230	15‡
India	1985	220 (25)	93 (25)	86 (25)	110	10	40	160	54
Asia		753 (25)	133 (25)	545 (25)	385	15	280	680	23
Global		1324 (25)	136 (25)	597 (25)	675	15	300	990	7

*Central value of biofuel consumption for cooking and BC emissions.   †Central value of percent contribution of BC from biofuel combustion.   ‡Lower and upper bound estimates of biofuel consumption for cooking and BC emissions at 95% confidence interval. The ranges are from two different studies (8, 25).   §Asia and global numbers include biofuel consumption for cooking and space heating, excluding the amount used in industry.
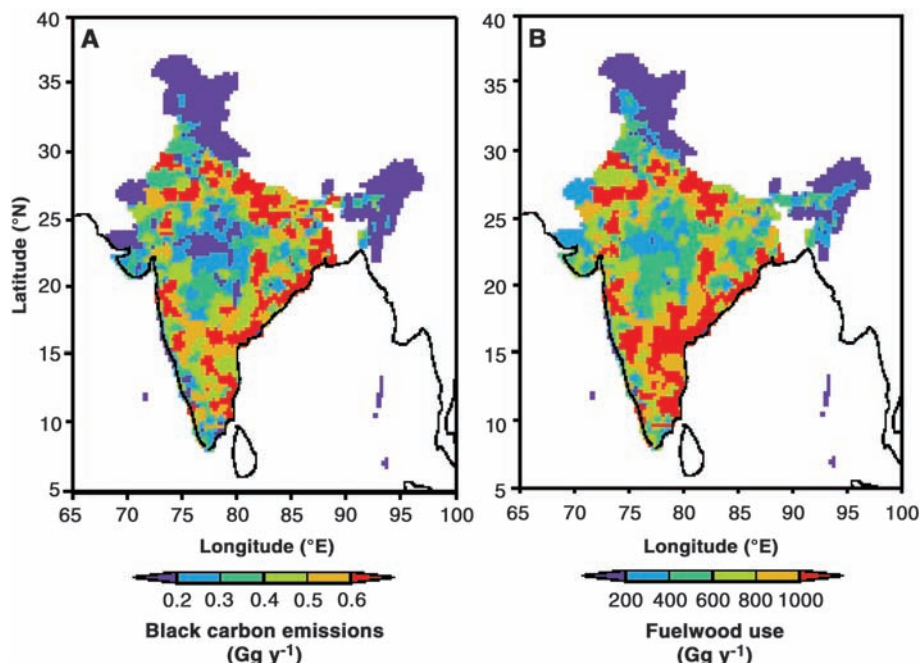
Fig. 1. Spatial distribution of (**A**) black carbon emissions (Gg year⁻¹) from biofuel combustion and (**B**) fuelwood use (Gg year⁻¹) in India.

the relative contributions to total OC emissions from fossil fuel, open burning, and biofuel combustion are 13%, 43%, and 44%, respectively.

Between 1985 and 1995, the reduction in the relative contribution of biofuels to Indian BC emissions (Table 1) resulted not from a reduction in this source, but from an increase in BC emissions from fossil fuel combustion during this period. Biofuel BC emissions from India have essentially remained unchanged during the 1985–1995 period (Table 1), implying that cleaner cooking technologies have been introduced very slowly. Other estimates of 207 to 425 Gg year⁻¹ BC and 484 to 2105 Gg year⁻¹ OC emissions from biofuel combustion in India (8, 11, 12) differ from our findings in that they use calculated emission factors, highly uncertain per capita biofuel usage in India, and an assumed 100% rural user population for all three biofuel types, thus making direct comparisons untenable.

High BC concentrations and atmospheric absorption measured during the Indian Ocean Experiment (INDOEX) have been related through trajectory analysis to source regions in the Indo-Gangetic plain, central/east coast, and south India (1, 27). The potential contribution of biofuel emissions was examined by estimating spatial distributions of biofuel combustion and related BC emissions, using district-level biofuel user populations (28) and the emission factors reported here. The emission rates of BC from biofuels (Fig. 1A) and use of wood as biofuel (Fig. 1B) were both high in regions from which highly absorbing

aerosols had originated during INDOEX. This suggests that the combustion of biofuels, especially wood, is a potentially significant source of atmospheric BC and related climate effects (5) in south Asia. The large radiation perturbations from aerosols (4) and the resulting potential changes in tropical rainfall (5) would have important implications for agricultural productivity and the economy of the region.

An analysis of the climate response of soot emissions from fossil fuel and biofuel combustion has suggested that control of soot, in addition to greenhouse gases, is an important measure to slow global warming, especially on short time scales (6, 7). Our results suggest that biofuel combustion could significantly affect atmospheric BC concentrations in the south Asian region. The climate effects of biofuel combustion aerosols have been combined with the effects of open biomass burning in the scientific consensus reports of the Intergovernmental Panel on Climate Change (29). We suggest that biofuel combustion needs to be addressed as a distinct source, and that cleaner cooking technologies not only could yield significant local health and air quality benefits but also could have an important role in climate change mitigation in the south Asian region.

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30. Supported by grants from the Indian Space Research Organization–Geosphere Biosphere Program (ISRO-GBP) and the Center for Clouds, Chemistry and Climate (C4), University of California, San Diego. Additional support (A.H.M. and A.E.-F.) was provided by National Institute of Environmental Health Sciences grant 5P30 ES07048 to the Southern California Environmental Health Sciences Center and by U.S. EPA grants R827352-01-0 and CR-82805901 to the Southern California Particle Center and Supersite. We thank M. Hakami for chemical analysis assistance. The late Prof. Glen R. Cass (California Institute of Technology and Georgia Institute of Technology) was instrumental in the initiation of this work.