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Emissions and Climate-Relevant Optical Properties of Pollutants Emitted from a Three-Stone Fire and the Berkeley-Darfur Stove Tested under Laboratory Conditions

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Supporting Information

ABSTRACT: Cooking in the developing world generates pollutants that endanger the health of billions of people and contribute to climate change. This study quantified pollutants emitted when cooking with a three-stone fire (TSF) and the Berkeley-Darfur Stove (BDS), the latter of which encloses the fire to increase fuel efficiency. The stoves were operated at the Lawrence Berkeley National Laboratory testing facility with a narrow range of fuel feed rates to minimize performance variability. Fast (1 Hz) measurements of pollutants enabled discrimination between the stoves' emission profiles and development of woodsmoke-specific calibrations for the aethalometer (black carbon, BC) and DustTrak (fine particles, PM_{2.5}). The BDS used 65 ± 5% (average ±95% confidence interval) of the wood consumed by the TSF and emitted 50 ± 5% of the carbon monoxide emitted by the TSF for an equivalent cooking task, indicating its higher thermal efficiency and a modest improvement in combustion efficiency. The BDS reduced total PM_{2.5} by 50% but achieved only a 30% reduction in BC emissions. The BDS-emitted particles were, therefore, more sunlight-absorbing: the average single scattering albedo at 532 nm was 0.36 for the BDS and 0.47 for the TSF. Mass emissions of PM_{2.5} and BC varied more than emissions of CO and wood consumption over all tests, and emissions and wood consumption varied more among TSF than BDS tests. The international community and the Global Alliance for Clean Cookstoves have proposed performance targets for the highest tier of cookstoves that correspond to greater reductions in fuel consumption and PM_{2.5} emissions of approximately 65% and 95%, respectively, compared to baseline cooking with the TSF. Given the accompanying decrease in BC emissions for stoves that achieve this stretch goal and BC's extremely high global warming potential, the short-term climate change mitigation from avoided BC emissions could exceed that from avoided CO₂ emissions.



INTRODUCTION

Three billion people use open fires or rudimentary cookstoves to burn wood, coal, dung, and agricultural waste for domestic cooking. These traditional forms of cooking are generally fuel inefficient because most of the fire's heat escapes to the environment. Moreover, these inefficient fires are grossly polluting and negatively impact indoor air quality.¹ A recent assessment of the global burden of disease found that 4 million premature deaths each year can be attributed to household air pollution from solid fuels, including 500,000 annual deaths from childhood pneumonia.²

In addition to these health impacts, the widespread use of traditional cookstoves throughout the developing world contributes to global and regional climate change. Emissions of carbon dioxide (CO₂) from burning nonrenewable biomass and emissions of black carbon (BC), the fraction of submicrometer-sized soot that strongly absorbs solar radiation, are of notable concern for their impact on climate. A recent study reaffirms the importance of BC as an anthropogenic atmospheric warming pollutant that is second only to CO₂,³

and residential biomass combustion accounts for ~30% of global anthropogenic BC emissions.⁴ Regional climate effects of BC emissions include changes in precipitation⁵ and accelerated melting of Himalayan glaciers,⁶ the latter of which has been directly linked to emissions from biofuel cooking.⁷

Efforts have been made to develop and disseminate cleaner cookstoves that reduce the local impact on biofuel resources, hardship of women, and exposure to harmful smoke, while simultaneously helping to mitigate climate change through emission reductions.^{8–13} Many of these endeavors are now united behind the Global Alliance for Clean Cookstoves (GACC), an initiative to “foster the adoption of clean cookstoves and fuels in 100 million households by the year 2020.”¹⁴ Performance tiers have been proposed as a means of ranking cookstoves, according to which the highest tier (tier 4)

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would reduce fuel use by 65–70% and PM_{2.5} emissions by 90–95% compared to baseline cooking with a traditional three-stone fire (TSF).^{15–17}

This study compared the performance of a traditional wood-burning TSF, an arrangement of three large stones with irregular shape supporting a pot over an open and unvented fire,^{18,19} and the Berkeley-Darfur Stove (BDS). Both stoves are shown in Figure S1 in the Supporting Information (SI). The BDS was designed and tested to reduce the amount of wood needed for cooking through increased thermal efficiency compared to cooking over a TSF.⁹ The physical design of the BDS is based on India's Tara Stove but modified for lower cost, longer life, easier assembly, the cultural cooking requirements of Darfuri women, and the windy conditions of the region.⁹ As with the Tara Stove, the BDS contains the fire in a low-emissivity stainless steel firebox, thereby focusing the radiative heat of the fire to the pot. Additional details about the humanitarian need that motivated the development of the BDS and features of the BDS design are provided in the SI.

While intended as a more fuel-efficient replacement of the TSF, the present study examined the BDS for the potential cobenefit of reduced pollutant emissions. With air pollution and associated environmental impacts in mind, this study quantified the emissions of carbon monoxide (CO), fine particulate matter (PM_{2.5}), and BC, as well as several climate-relevant particle optical properties and fuel consumption. The BDS design elements for increased fuel efficiency are common to stoves developed for use in other regions of the world, including a metal combustion chamber, a raised grate upon which fires are maintained for improved airflow, and a tapered wind collar.^{19,20} As such, the results of this study may be relevant to other wood-burning, natural-draft stoves with similar design elements.

The cooking tasks in this study were performed by two well-trained scientists under conditions of narrowly constrained fuel-feed rates. Pollutant concentrations were measured with high time resolution to enable a comparison of the emissions from each stove throughout cooking tests. The time-resolved measurements also enabled the development of woodsmoke-specific calibrations of the analyzers used to measure BC and PM_{2.5} concentrations.

METHODS

Testing Facility and Measurements. All measurements were made at the cookstove testing facility at Lawrence Berkeley National Laboratory. Cooking was carried out on a platform underneath a large plenum that captured and diluted the stove's emissions. Details of this facility, including the sampling setup, airflow measurements, and instrument specifications, are in the SI (Figure S2 and Table S1).

The concentrations of CO, CO₂, BC, and PM_{2.5} as well as particle absorption and scattering coefficients (b_{abs} and b_{scat}) in diluted exhaust were measured at a temporal resolution of 1 Hz. CO and CO₂ concentrations were measured in a single instrument using nondispersive infrared absorption spectroscopy, which was calibrated before and after each test. PM_{2.5} and BC concentrations were measured using a DustTrak and an aethalometer, respectively. Particle absorption and scattering coefficients at a wavelength of 532 nm were measured using a custom-made photoacoustic absorption spectrometer (PAS)²¹ equipped with a reciprocal nephelometer.²² The calibration of the PAS was verified prior to this study using ammonium sulfate and soot particles, as described by Arnott et al.²³ A

three-wavelength particle soot-absorption photometer (PSAP, not shown in Figure S2, SI) was used in a subset of tests to simultaneously measure particle light-absorption coefficients at 467, 530, and 660 nm.²⁴

Woodsmoke Specific Calibrations for BC and PM_{2.5}. Time-integrated concentrations of BC and PM_{2.5} were periodically measured using quartz and Teflon filters in order to develop calibration equations specific to woodsmoke for the aethalometer and DustTrak, as described in the SI. The BC concentrations reported by the aethalometer were adjusted to correct for a known sampling artifact that causes the aethalometer's response to diminish as its filter darkens during sampling (see eq S1 and Figure S3, SI).²⁵ The PM_{2.5} concentrations measured with the DustTrak were also adjusted to a woodsmoke specific calibration, as per the manufacturer's recommendation (see eq S2 and Figure S4, SI).

Optical Properties. Extinction-weighted aerosol single scattering albedo (SSA), the fraction of incident light that is scattered rather than absorbed by particles, was calculated from PAS measurements of b_{abs} and b_{scat} (in units of Mm⁻¹) using eq 1, where t_0 and t_f are the test start and end times, and b_{ext} is particle extinction ($b_{\text{ext}} = b_{\text{abs}} + b_{\text{scat}}$). Mass-specific absorption (MAE) and scattering (MSE) efficiencies (in units of m² g⁻¹) were calculated from measured b_{abs} , b_{scat} , and concentrations of PM_{2.5} and BC using eqs 2–4. The relative humidity measured inside the PAS was typically 10%. Therefore, the sampled particulate matter is presumed to have been dry, such that b_{abs} was unaffected by evaporation of water from particle surfaces and b_{scat} was not enhanced by water-increased particle cross-section.²⁶ The absorption Ångström exponent (AAE), a measure of the variation in light-absorption with wavelength (i.e., absorption selectivity),²⁷ was calculated from PSAP measurements of b_{abs} at three wavelengths (see eq S3, SI).

$$\text{extinction-weighted SSA (532 nm)} = \frac{\int_{t_0}^{t_f} \text{SSA}(t) b_{\text{ext}}(t) dt}{\int_{t_0}^{t_f} b_{\text{ext}}(t) dt} \quad (1)$$

$$\text{MAE}_{\text{PM}_{2.5}} (532 \text{ nm}) = \frac{b_{\text{abs}}}{[\text{PM}_{2.5}]} \quad (2)$$

$$\text{MAE}_{\text{BC}} (532 \text{ nm}) = \frac{b_{\text{abs}}}{[\text{BC}]} \quad (3)$$

$$\text{MSE}_{\text{PM}_{2.5}} (532 \text{ nm}) = \frac{b_{\text{scat}}}{[\text{PM}_{2.5}]} \quad (4)$$

Test Protocol. The BDS and TSF are compared in this study using the same constantly attended cooking test, which featured the boiling of 2.5 L of water in a 2.3 kg metal Darfur pot. This modified cold-start water boiling protocol was intended to simulate the cooking of assida, a porridge made of flour and water that is a staple food in Darfur.¹⁸ This doughy dish is traditionally cooked over a high-powered fire to bring the water to a boil and then simmered over lower heat until the water is completely absorbed.⁹ In this test, a wood fire was ignited by burning one sheet of crumpled newspaper with smaller pieces of kindling. The fire was then built up by addition of wood to heat the water from an initial temperature of 21 ± 4 °C (± 1 standard deviation) to 100 °C. Once the water reached a boil, fuelwood was added at a rate sufficient to keep the water at a simmer above 94 °C for 15 min. The fuel-

feed rates for both stoves were similar and are summarized in the results below. The fire tender arranged wood pieces to maintain the fire and on rare occasion used a bellows to blow on the fire when it prematurely extinguished. After the 15 min simmer period, emissions measurements were stopped and the mass of wood remaining was immediately measured. (This test, therefore, does not include a characterization of the pollutant emissions that occur during the exclusively nonflaming, smoldering conditions of fires that extinguish naturally.)

The mass and moisture content of each piece of wood was measured using a calibrated analytical balance (5 kg capacity, 0.1 g resolution) and a moisture meter (6–40% moisture range, 0.1% resolution). The equivalent dry mass of each piece of wood was then calculated (see eq S4, SI). Soft (pine and fir) and hard (oak) woods were used in an equal number of tests with both stove types. While these specific woods are not identical to those used in Darfur, they serve as a substitute for the two densities of wood that are commonly burned in the displacement camps and follow the practice of previous laboratory testing during the development of the BDS.^{9,18} Soft wood pieces were saw-cut to an approximate length of 15 cm with a square cross-section of approximately 4 cm² and hard wood pieces were hatchet-cut to a similar size but irregular shape. The average (± 1 standard deviation) moisture content and dry mass of individual pieces of soft wood ($9 \pm 1\%$ and 20 ± 9 g) and hard wood ($10 \pm 1\%$ and 26 ± 13 g) were similar to each other and were the same for TSF and BDS tests (see Table 2 and Table S6, SI).

Pollutant Emission Calculations. Emission factors were computed as mass of pollutant emitted per unit mass of wood burned using the carbon balance method²⁸

$$EF_p = 10^3 \left[\frac{\Delta[P]}{\Delta[CO] + \Delta[CO_2]} \right] w_c \quad (5)$$

where EF_p is the emission factor (g emitted per kg of fuel burned) for pollutant P, $\Delta[P]$ is the increase in the concentration of pollutant P ($\mu\text{g m}^{-3}$) above background levels, $\Delta[CO]$ and $\Delta[CO_2]$ are the increases in the concentrations of CO and CO₂ ($\mu\text{g of carbon m}^{-3}$) above background levels, and w_c is the fraction of carbon in wood. Multiplying by w_c relates total carbon emissions in the fire (mainly in the form of CO and CO₂) to the carbon content of wood. The value of w_c was assumed to be 0.5 (g carbon per g of wood).²⁹ Background levels of all species except CO₂ were negligible compared to concentrations measured in the woodsmoke in this study. Background CO₂ concentrations were measured prior to the ignition of each fire.

Fuel-based emission factors were computed for each cooking test using test-average pollutant concentrations. The total mass (g) of a pollutant emitted in each test was calculated as the product of the test-average fuel-based emission factor and the measured mass of wood burned during the test. Pollutant emissions expressed in terms of μg emitted per second were computed from 1 Hz measurements

$$IEF_p = Q\Delta[P] \quad (6)$$

where IEF_p is the “instantaneous” emission factor for pollutant P, $\Delta[P]$ is defined above, and Q is the flow rate of air ($\text{m}^3 \text{s}^{-1}$) into the exhaust plenum. Q was measured at 1 Hz, as noted in the SI.

RESULTS

The data presented below are the result of 41 tests, 21 with the BDS and 20 with the TSF, conducted after completing a comparable number of preliminary tests intended to establish consistency in fire tending methods and emissions measurements. The instrumentation described above and listed in Table S1, SI was used in all tests, except the PSAP, which was available only during preliminary testing. AAE values derived from the PSAP data are the result of 15 preliminary tests. All average values are reported with corresponding 95% confidence intervals, except where noted.

Cooking Time and Wood Consumption. The time-resolved water temperature and the mass of (dry) wood consumed during BDS and TSF tests are shown in Figures S6 and S7, SI. Tests with the BDS were completed in $74 \pm 5\%$ of the time and with $65 \pm 5\%$ of the wood required for the TSF. Tests with both stoves were conducted with a narrow range of fuel-feed rates ($12.8 \pm 0.4 \text{ g min}^{-1}$ for the BDS and $14.9 \pm 0.6 \text{ g min}^{-1}$ for the TSF), illustrating intended consistency from test to test. Fires were maintained with a higher fuel-feed rate to bring the water to boil than during the subsequent 15 min simmering period. Whereas the wood addition rate was essentially the same during the preboil, higher power portion of BDS tests ($20.1 \pm 0.9 \text{ g min}^{-1}$) and TSF tests ($19.6 \pm 1.2 \text{ g min}^{-1}$), the average water heating rate was about 1.6 times higher when cooking with the BDS compared to cooking with the TSF (Figure S6, SI), which is indicative of the higher thermal efficiency of the BDS. The wood addition rate during the postboil, lower power portion of TSF tests ($7.1 \pm 0.7 \text{ g min}^{-1}$) was 23% higher than during BDS tests ($5.5 \pm 0.5 \text{ g min}^{-1}$), indicative of the fact that the TSF required more wood than the BDS to maintain a simmer. Also, the water heating rate and wood mass required to complete tests were less variable from test to test when cooking with the BDS, as illustrated (for wood mass) in Figure S7, SI, by a narrower distribution and smaller confidence interval.

Figure 1 shows normalized cumulative emissions of CO₂, based on instantaneous emission factors calculated with eq 6 (i.e., $\mu\text{g s}^{-1}$) and averaged over all tests with the BDS and TSF. The shaded bands indicate the 95% confidence intervals about the mean values. The cumulative mass emission normalized to the total mass emitted during each test ranges from 0 to 1 on the vertical axis, and time is shown on the horizontal axis normalized to the total duration of the test. The average point at which the water reached a boil (and the simmering portion of the test commenced) is indicated.

Halfway through tests with the BDS, which was usually close to the time when the water started to boil, approximately 80% of the total wood had been added and 60% of total mass of CO₂ had been emitted. The CO₂ emission rate was higher during the first half of BDS tests because the fuel-feed rate was higher when bringing the water to boil (i.e., higher power) than when simmering the water for 15 min (i.e., lower power).

Time-Resolved Pollutant Emissions. Time-resolved emissions of CO, PM_{2.5}, and BC are presented in Figure 1. Table 1 reports emissions averaged over five segments of the cooking test: (1) the first 3 min, which captures the lighting of the fire; (2) fire ignition to water boil, including the first 3 min; (3) the 15 min simmer after the water reached a boil; (4) the last 3 min of the 15 min simmer, when the fire was minimally attended by adding wood only if needed to maintain water temperature above 94 °C; and (5) the entire test period.

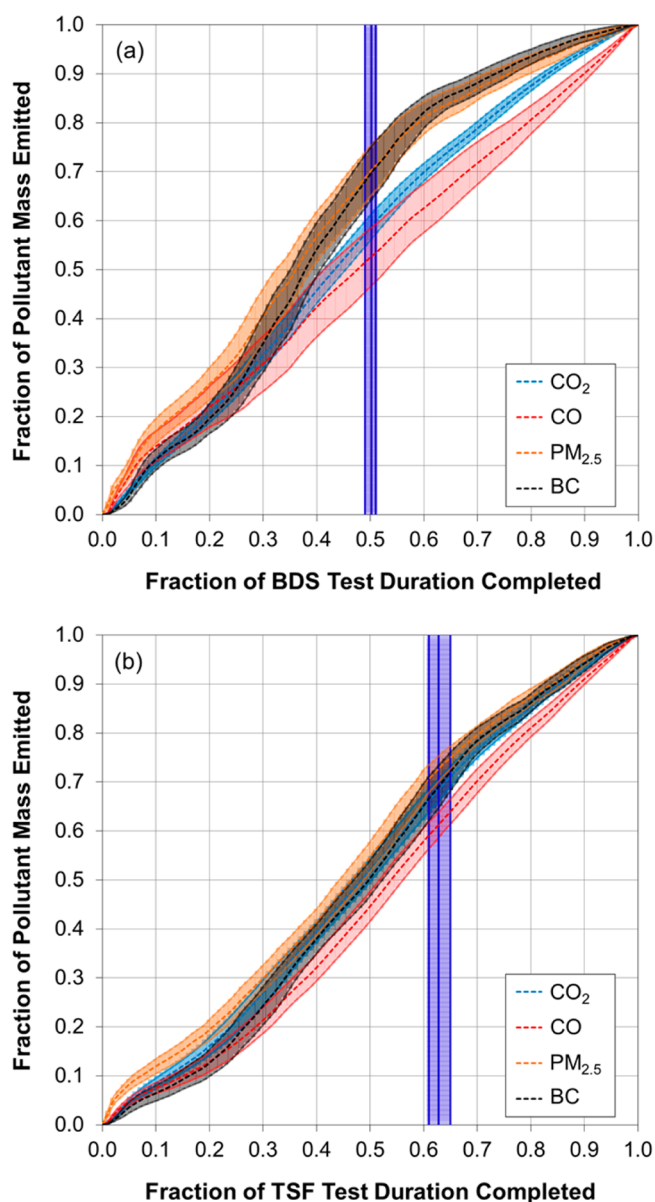


Figure 1. (a) Normalized cumulative emission of pollutants averaged over all BDS tests. (b) Normalized cumulative emission of pollutants averaged over all TSF tests. The vertical solid blue line indicates the average point when the water began to boil and the simmering portion of the test started. The shaded bands represent the 95% confidence intervals around the average values.

As illustrated in Figure 1, the patterns of pollutant emissions during BDS and TSF tests were different. Notably, in BDS tests, PM_{2.5} and BC emission rates (g s^{-1}) markedly increased relative to the CO₂ emission rate prior to boiling (at about a quarter of the way into the test) and decreased relative to the CO₂ emission rate after boiling (after 60% of the test was completed). In contrast, the emission rates of PM_{2.5} and BC during TSF tests were generally proportional to the CO₂ emission rate and did not vary as much as they did in BDS tests. Drawing from the normalized emission rates reported in Table 1, PM_{2.5} and BC emission rates in BDS tests were 2.4 times higher prior to boiling than after boiling (1.41/0.60 for PM_{2.5} and 1.40/0.59 for BC). The distinction was much smaller for TSF tests: PM_{2.5} and BC emission rates were 1.6 and 1.3 times higher preboiling than postboiling. We infer from these

different emission patterns that the combustion conditions affecting PM_{2.5} and BC emissions were more distinct during the preboiling and simmering phases of cooking with the BDS than with the TSF. For example, relatively high BC emission rates are typical of fires that are predominantly flaming combustion rather than smoldering combustion.³⁰ These observations indicate that efforts to design a new version of the BDS that emits less BC and PM_{2.5} may be most effective if focused on suppressing particles produced during the high power portion of cooking events.

Compared to PM_{2.5} and BC, the CO emission rates pre- and postboiling were less distinct. During BDS tests, the CO emission rate (g s^{-1}) was 1.17 times higher preboiling. During TSF tests, the CO emission rate was 1.04 times higher postboiling (Table 1). Thus, the combustion conditions that distinguished pre- and postboiling emission rates of PM_{2.5} and BC had less of an influence on the CO emission rate.

Time-resolved emissions measurements also illustrate that the ignition portion of tests (i.e., the first 3 min) often exhibited the highest PM_{2.5} emission rate, and these events that represented 10% of the test duration amounted to an average of 17% of total test PM_{2.5} emissions for the BDS. The average PM_{2.5} emission rates (g s^{-1}) during the first 3 min of tests were nearly 5 and 3 times higher than those during the last 3 min for the BDS and TSF, respectively (Table 1). Similarly, the average BC emission rate during the ignition portion of BDS tests was almost 6 times higher than during the last 3 min. In contrast, BC emission rates during both segments were equal in TSF tests.

Since fires were extinguished after the 15 min simmering period, this study did not capture the purely smoldering emissions that occur when fires naturally die out. Fire extinguishing reflects the cooking practice in Darfur, where fires are quickly quenched to conserve wood rather than allowing fires to smolder. Emissions of CO would likely be high, and emissions of BC likely low, for both stoves during late-stage smoldering.³¹

Time-Integrated Pollutant Emissions. Average pollutant emission factors and total mass emissions for each cooking test are reported in Tables S4 and S5, SI. These results are based on average pollutant concentrations measured during each test, as reported in Tables S2 and S3, SI. Figure 2 compares the total masses of CO, PM_{2.5}, and BC emitted during each BDS and TSF test. Emission factors and mass emissions averaged over all BDS and TSF tests and reported below are summarized in Table 2.

Of these three pollutants, the clearest distinction between the BDS and TSF is observed for CO. Though mass emissions of CO varied by almost a factor of 2 for each stove (e.g., ~10 to 20 g for the BDS), the most CO emitted during any one BDS test was less than the CO emitted in the lowest-emitting TSF test (Figure 2a and b). The average fuel-based CO emission factor (g kg^{-1}) during tests with the BDS was $75 \pm 6\%$ of that for the TSF. The distinction between the stoves is larger when factoring in the higher fuel efficiency of the BDS: on average the BDS emitted $50 \pm 5\%$ of the CO mass (g) emitted by the TSF for the same cooking task.

The average PM_{2.5} emission factor (g kg^{-1}) during tests with the BDS was $79 \pm 16\%$ of that for the TSF. Including the higher fuel efficiency of the BDS, on average the BDS emitted $52 \pm 11\%$ of the PM_{2.5} emitted by the TSF. Figure 2a illustrates that the mass of PM_{2.5} emitted during 45% of TSF tests (i.e., 9 out of 20) exceeded the mass of PM_{2.5} emitted during all tests

Table 1. Summary of Emissions of CO₂, CO, PM_{2.5}, and BC by Test Segment for the BDS and TSF

stove	test segment	avg duration (min)	normalized avg duration ^a	normalized avg emission rate ^b				normalized avg mass emissions ^c			
				CO ₂	CO	PM _{2.5}	BC	CO ₂	CO	PM _{2.5}	BC
BDS (n = 21)	first 3 min	3.0	0.10	1.04	1.46	1.68	1.19	0.10	0.14	0.17	0.12
	pre-boil	15.3	0.50	1.16	1.08	1.41	1.40	0.59	0.54	0.71	0.71
	post-boil	15.0	0.50	0.84	0.92	0.60	0.59	0.41	0.46	0.29	0.29
	last 3 min	3.0	0.10	0.55	1.04	0.37	0.20	0.05	0.10	0.04	0.02
	entire test	30.3	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
TSF (n = 20)	first 3 min	3.0	0.07	0.88	0.82	1.36	0.65	0.06	0.06	0.10	0.05
	pre-boil	26.0	0.63	1.07	0.99	1.15	1.10	0.68	0.62	0.73	0.70
	post-boil	15.0	0.37	0.86	1.03	0.73	0.82	0.32	0.38	0.27	0.30
	last 3 min	3.0	0.07	0.64	0.92	0.50	0.65	0.05	0.07	0.04	0.05
	entire test	41.0	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

^aAverage duration of test segment normalized by average test duration. ^bSegment average instantaneous emission rate (g s⁻¹) normalized by the test average instantaneous emission rate. ^cSegment average mass emission (g) normalized by the test average mass emission.

with the BDS. Likewise, the mass of PM_{2.5} emitted during 38% of BDS tests (i.e., 8 out of 21) was less than that emitted during all tests with the TSF. As a result of substantial variation in emissions from one test to the next, the PM_{2.5} mass emissions varied by almost a factor of 4 (e.g., ~0.5 to 2.0 g PM_{2.5} for the BDS) and more than emissions of CO.

The average BC emission factor (g kg⁻¹) during tests with the BDS was 115 ± 20% of that with the TSF. Although the fuel-based BC emission factor was higher for the BDS, the smaller amount of fuel consumed by the BDS resulted in less BC emitted overall, such that the BDS emitted on average 71 ± 15% of the BC mass emitted by the TSF. BC emissions varied considerably from test to test and, compared to PM_{2.5} emissions, there was greater overlap in distributions of BC emissions from the BDS and TSF (Figures 2a and b).

Given these results for PM_{2.5} and BC, the emission ratio of BC/PM_{2.5} was larger for the BDS than the TSF (Figure S12, SI). Consequently, the MAE_{PM2.5} (532 nm) of emitted by the BDS (4.6 ± 0.3 m² g⁻¹) was higher than that emitted by the TSF (3.1 ± 0.4 m² g⁻¹). The MAE_{BC} (532 nm) of BC was quite similar for both stoves (9.8 ± 0.6 m² g⁻¹ for BDS and 9.0 ± 0.7 m² g⁻¹ for TSF). Consistent with these results, the PM_{2.5} emitted by the BDS had a lower SSA (Figure 2c and Table 2) and a lower AAE (Figure S14, SI) compared to the PM_{2.5} emitted by the TSF. The lower SSA (average of 0.36 ± 0.02 for BDS versus 0.47 ± 0.03 for TSF) means that BDS smoke would absorb a greater fraction of incident solar radiation than TSF smoke: 64% versus 53%, respectively, based on the SSA complement (i.e., 1-SSA). The lower AAE (1.26 for BDS versus 1.51 for TSF) means that, on the whole, the BDS smoke would absorb solar radiation with less wavelength selectivity (i.e., the smoke is blacker) than the TSF smoke.^{27,32}

Effect of Wood Type. Table S6 (SI) presents wood type-specific averaged test results. Tests with soft wood were completed in about 90% of the time and with 90% of the wood compared to tests with hard wood. The relative efficiency of the BDS and the TSF, measured in time required and dry wood consumed in completing the cooking task, was essentially the same for both wood types.

There were some differences in emissions that appeared to be linked to wood type, but regardless of the type used, total mass emissions of CO, PM_{2.5}, and BC were lower when cooking with the BDS than the TSF. This is evidenced by BDS/TSF emission ratios (see the last four columns of Table S6, SI) that are less than 1 for each of these pollutants. With both stoves, one standout was the higher emission of PM_{2.5} and

BC when cooking with pine, which we suspect was due to high sap content, despite an attempt to exclude pinewood with visible surface sap.

DISCUSSION

Emissions Reduction Potential. In this study, the BDS used 65 ± 5% of the wood consumed by the TSF for an equivalent water boiling task, which is reflective of the higher thermal efficiency of the BDS than the TSF. The incrementally larger reductions in emitted CO and PM_{2.5} mass (50 ± 5% of the CO and 52 ± 11% of the PM_{2.5} emitted by the TSF) indicate that the BDS design also led to a modest improvement in combustion efficiency. By comparison, Jetter et al.²⁰ recently reported better fuel efficiency and emissions performance than measured in the current study: relative to the TSF, the BDS consumed 43% of the wood and emitted 40% of the CO and 36% of the PM_{2.5} (Table S7, SI). This difference may be related to fuel-feed rate, which was more than two times higher for the TSF than the BDS in the study of Jetter et al. and similar for the two stoves in the current study. Jetter et al. also found that the BDS had CO and PM_{2.5} emissions comparable to other wood-burning, natural-draft stoves.²⁰ These results indicate the potential for reducing environmental and health impacts associated with wood scavenging, firewood costs, and exposure to smoke in the developing world. As of early 2014, more than 37000 stoves have been distributed in Darfur with an additional 10000 planned in the next 12 months.³³

The performances of the BDS and many other stoves fall short of the 65–70% fuel savings and 90–95% PM_{2.5} emission reduction targets relative to baseline cooking established by the international community for tier 4 cookstoves.^{16,17,20} However, these targets can be achieved through additional design modifications.³⁴ In particular, stoves that promote the mixing of air in the flame zone—either naturally via stove design or forced via the addition of a fan—can effectually reduce both gaseous and particulate pollutants by an order of magnitude.²⁰ At the time of this publication, development of an advanced version of the BDS for substantially larger emissions reductions is underway.

Climate Implications. With average extinction-weighted SSAs of 0.36 and 0.47, far below the climate cooling-to-warming threshold of about 0.85,^{35,36} PM_{2.5} emissions from both the BDS and TSF are climate warming. Based on these SSA values, BDS-emitted particles absorb 20% more light (at 532 nm) than particles emitted by the TSF ($[1 - 0.36]/[1 - 0.47] = 1.2$). While the particles emitted by the BDS were 20%

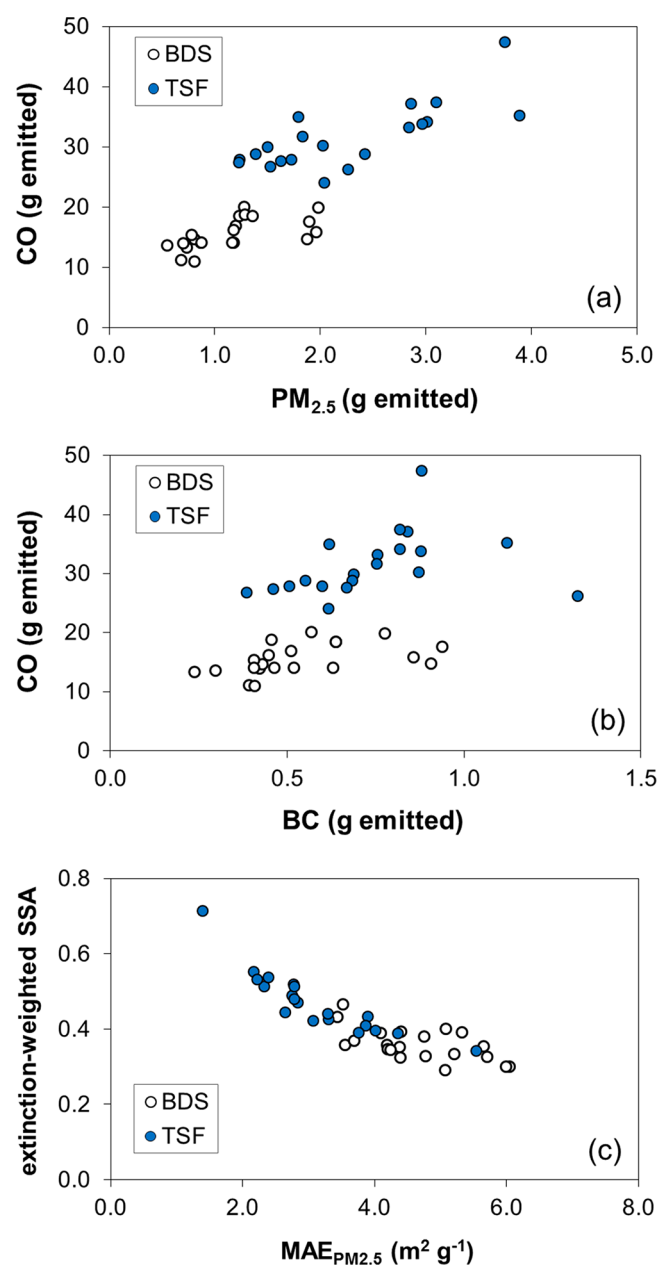


Figure 2. From the 21 BDS and 20 TSF tests: (a) total mass emissions of PM_{2.5} and CO; (b) total mass emissions of BC and CO; and (c) the relationship between the calculated MAE_{PM_{2.5}} and extinction-weighted SSA values of the emitted particles, measured at 532 nm.

more light-absorbing on a per gram basis than those emitted by the TSF, the BDS emitted 50% less PM_{2.5} and 30% less BC mass than the TSF. Considering both factors, BDS particulate matter emissions would likely have a smaller climate warming effect than TSF particulate matter emissions, on a per meal basis (assuming that our modified water boiling tests are a good surrogate for a meal). A more robust evaluation requires climate simulations, which could be conducted to study regional and global impacts of replacing traditional stoves with more efficient and lower emitting stoves on a large scale—including impacts of reduced emissions of BC and non-BC PM_{2.5}.

Carbon-trading schemes that quantify reduced CO₂ emissions for projects that replace traditional cooking methods with more efficient stoves may aid in their expanded

Table 2. Wood Consumed, Fuel-Based Pollutant Emission Factors, Total Mass of Pollutants Emitted, and Particle Optical Properties (at 532 nm), Averaged over All BDS and TSF Tests Conducted, with 95% Confidence about the Means Noted

	BDS (<i>n</i> = 21)	TSF (<i>n</i> = 20)	BDS/TSF ratio
wood:			
moisture content	0.09 ± 0.01	0.09 ± 0.01	1.00
dry mass (g test ⁻¹)	368 ± 15	564 ± 32	0.65
emission factors:			
CO ₂ (g kg ⁻¹)	1767 ± 4	1745 ± 5	1.01
CO (g kg ⁻¹)	42 ± 3	56 ± 3	0.75
PM _{2.5} (g kg ⁻¹)	3.1 ± 0.5	3.9 ± 0.4	0.79
BC (g kg ⁻¹)	1.5 ± 0.2	1.3 ± 0.1	1.15
BC/PM _{2.5}	0.48 ± 0.03	0.35 ± 0.04	1.37
total mass emitted:			
CO ₂ (g test ⁻¹)	650 ± 25	985 ± 57	0.66
CO (g test ⁻¹)	16 ± 1	32 ± 2	0.50
PM _{2.5} (g test ⁻¹)	1.2 ± 0.2	2.3 ± 0.3	0.52
BC (g test ⁻¹)	0.5 ± 0.1	0.7 ± 0.1	0.71
emitted particle optical properties:			
MAE _{PM_{2.5}} (m ² g ⁻¹)	4.6 ± 0.3	3.1 ± 0.4	1.48
MAE _{BC} (m ² g ⁻¹)	9.8 ± 0.6	9.0 ± 0.7	1.09
MSE _{PM_{2.5}} (m ² g ⁻¹)	2.6 ± 0.2	2.7 ± 0.1	0.96
SSA	0.36 ± 0.02	0.47 ± 0.03	0.77
1-SSA	0.64 ± 0.02	0.53 ± 0.03	1.21

dissemination.^{37,38} While having the potential climate mitigation benefits of reduced CO₂ emissions, the current study illustrates that efficient stoves also have the potential to substantially reduce particle emissions. This is significant because the global warming potential (GWP) of BC has been estimated as 910 for a 100 year time scale and as 3200 for a 20 year time scale.³ As noted above, compared to all anthropogenic climate forcing agents in the atmosphere, BC emissions exert the second largest climate forcing and cookstoves are a major source of BC.^{3,7} Given that BC is a very short-lived species relative to CO₂ with a characteristic lifetime of weeks versus centuries, it is likely that its immediate climate impact is greater than is reflected by GWP on even a 20 year time scale and would be quickly realized.

The contribution to near-term climate change mitigation from a 95% reduction in BC emissions could be larger than that from a 65% reduction in CO₂ emissions, which are consistent with the fuel efficiency and PM_{2.5} emission targets for the highest performance tier of biomass stoves.^{16,17} For instance, reducing the average masses of BC (0.7 g, Table 2) and CO₂ (1000 g, Table 2) emitted during a TSF cooking event in this study by 95 and 65%, respectively, would eliminate the addition of 0.67 g of BC and, assuming nonrenewable biomass is used, 650 g of CO₂ to the atmosphere. Multiplying by its 20 year GWP of 3200 suggests that the short-term climate mitigation due to the elimination of 0.67 g of BC would be approximately 3 times larger than the mitigation due to the elimination of 650 g of CO₂. The relative benefit of reduced BC is even larger if some of the biomass is renewable. Therefore, changes to particle emissions and their climate relevant optical properties warrant consideration along with reductions of CO₂ emissions when examining the potential climate impacts of high performance cookstoves.

Emissions Variability. A number of recent studies have demonstrated that the efficiency and emissions of cookstoves

are highly variable and that stove performance in the field may be quite different than it is in the laboratory.^{39–41} These lab-field performance differences are likely due, at least in part, to the disparity in fire tending: cooking in the laboratory is likely to be well attended by a technician in a controlled environment and cooking in the field is likely not as closely tended and performed in a relatively uncontrolled environment. Even in the laboratory, stove test results may show a high degree of variability for a variety of reasons, including the frequency of fuel-feeding (as mentioned above) and characteristics of each piece of fuelwood, such as the relative amounts of bark and sap as well as the surface to volume ratio.⁴² To minimize variability in results owing to variability in the fuel-feeding rate, the latter was well constrained from test to test in the current study. The coefficient of variation of fuel-feeding rate (i.e., the standard deviation divided by the mean) was 8% for the BDS and 9% for the TSF.

In our laboratory study under these conditions, PM_{2.5} and BC mass emissions varied by a factor of 4–5 over all tests, considerably more than emissions of CO, which varied by a factor of about 2, and fuel use, which varied by less than a factor of 2 over all tests. These results may be an indication that the noted discrepancies between lab and field may be more of an issue for PM_{2.5} and BC emissions than for CO emissions and efficiency performance. Fuel consumption and pollutant emissions results varied more among TSF tests than among BDS tests. The greater consistency in BDS results may be attributable to its design, which may promote a more stable fire throughout a cooking task compared to the TSF.

Typical laboratory testing relies on three replicate tests of a stove, but due to wide variability in stove testing results, such a small sample size can misrepresent a stove's mean performance. This important issue is the subject of a separate analysis of the data set provided by the current study, which has been published elsewhere.⁴² That analysis found that on the order of 10 replicate tests are necessary to accurately describe each stove's performance; fewer would be misleading, and a greater number of tests are recommended for field testing in a less controlled environment.⁴²

■ ASSOCIATED CONTENT

📄 Supporting Information

Additional details about the stoves, testing facility, and sampling designs, in addition to supplementary results, as specified in the manuscript. This material is available free of charge via the Internet at <http://pubs.acs.org/>.

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Notes

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