# **Environmental** Science & lechnology

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

Chelsea V. Preble,<sup>†,‡</sup> Odelle L. Hadley,<sup>‡,§</sup> Ashok J. Gadgil,<sup>†,‡</sup> and Thomas W. Kirchstetter<sup>\*,†,‡</sup>

<sup>†</sup>Department of Civil and Environmental Engineering, University of California, Berkeley, California 94720-1710, United States <sup>‡</sup>Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States

**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<sub>2.5</sub>). The BDS used 65  $\pm$  5% (average  $\pm$ 95% confidence interval) of the wood consumed by the TSF and emitted 50  $\pm$  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 BDSemitted 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  $CO_2$  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.<sup>1</sup> 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.<sup>2</sup>

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  $(CO_2)$  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  $CO_2^{3}$ 

and residential biomass combustion accounts for ~30% of global anthropogenic BC emissions.<sup>4</sup> Regional climate effects of BC emissions include changes in precipitation<sup>5</sup> and accelerated melting of Himalayan glaciers,<sup>6</sup> the latter of which has been directly linked to emissions from biofuel cooking.<sup>7</sup>

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.<sup>8–13</sup> 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."<sup>14</sup> Performance tiers have been proposed as a means of ranking cookstoves, according to which the highest tier (tier 4)

Received:	January 16, 2014
Revised:	March 30, 2014
Accepted:	March 31, 2014

would reduce fuel use by 65–70% and  $PM_{2.5}$  emissions by 90– 95% compared to baseline cooking with a traditional threestone fire (TSF).<sup>15–17</sup>

This study compared the performance of a traditional woodburning TSF, an arrangement of three large stones with irregular shape supporting a pot over an open and unvented fire,<sup>18,19</sup> 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.<sup>9</sup> As with the Tara Stove, the BDS contains the fire in a lowemissivity 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.<sup>19,20</sup> 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 welltrained scientists under conditions of narrowly constrained fuelfeed 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 woodsmokespecific 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, CO<sub>2</sub>, BC, and PM<sub>2.5</sub> 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 CO<sub>2</sub> concentrations were measured in a single instrument using nondispersive infrared absorption spectroscopy, which was calibrated before and after each test. PM<sub>2.5</sub> 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)<sup>21</sup> equipped with a reciprocal nephelometer.<sup>22</sup> The calibration of the PAS was verified prior to this study using ammonium sulfate and soot particles, as described by Arnott et al.<sup>23</sup> 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.<sup>24</sup>

**Woodsmoke Specific Calibrations for BC and PM**<sub>2.5</sub>. Time-integrated concentrations of BC and PM<sub>2.5</sub> 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).<sup>25</sup> The PM<sub>2.5</sub> 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 ( $b_{ext} = b_{abs} + b_{scat}$ ). Mass-specific absorption (MAE) and scattering (MSE) efficiencies (in units of  $m^2 g^{-1}$ ) were calculated from measured  $b_{abs}$ ,  $b_{scat}$ , and concentrations of PM<sub>2.5</sub> 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<sub>abs</sub> was unaffected by evaporation of water from particle surfaces and  $b_{scat}$  was not enhanced by water-increased particle cross-section.<sup>26</sup> The absorption Ångström exponent (AAE), a measure of the variation in light-absorption with wavelength (i.e., absorption selectivity),<sup>27</sup> was calculated from PSAP measurements of  $b_{abs}$  at three wavelengths (see eq S3, SI).

extinction-weighted SSA (532 nm) = 
$$\frac{\int_{t_0}^{t_f} SSA(t) \mathbf{b}_{ext}(t) dt}{\int_{t_0}^{t_f} \mathbf{b}_{ext}(t) dt}$$
(1)

$$MAE_{PM2.5} (532 \text{ nm}) = \frac{b_{abs}}{[PM_{2.5}]}$$
(2)

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

$$MSE_{PM2.5} (532 \text{ nm}) = \frac{b_{scat}}{[PM_{2.5}]}$$
(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.<sup>18</sup> 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.<sup>9</sup> 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  $21 \pm 4$  °C ( $\pm 1$  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-

#### **Environmental Science & Technology**

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  $4 \text{ cm}^2$  and hard wood pieces were hatchet-cut to a similar size but irregular shape. The average  $(\pm 1$  standard deviation) moisture content and dry mass of individual pieces of soft wood (9  $\pm$  1% and 20  $\pm$  9 g) and hard wood (10  $\pm$  1% and 26  $\pm$  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 method<sup>28</sup>

$$EF_{P} = 10^{3} \left[ \frac{\Delta[P]}{\Delta[CO] + \Delta[CO_{2}]} \right] w_{c}$$
(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 ( $\mu$ g m<sup>-3</sup>) above background levels,  $\Delta$ [CO] and  $\Delta$ [CO<sub>2</sub>] are the increases in the concentrations of CO and CO<sub>2</sub> ( $\mu$ g of carbon m<sup>-3</sup>) 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 CO<sub>2</sub>) to the carbon content of wood. The value of  $w_c$  was assumed to be 0.5 (g carbon per g of wood).<sup>29</sup> Background levels of all species except CO<sub>2</sub> were negligible compared to concentrations measured in the woodsmoke in this study. Background CO<sub>2</sub> 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  $\mu$ g emitted per second were computed from 1 Hz measurements

$$IEF_{P} = Q\Delta[P] \tag{6}$$

where IEF<sub>p</sub> is the "instantaneous" emission factor for pollutant P,  $\Delta$ [P] is defined above, and Q is the flow rate of air (m<sup>3</sup> s<sup>-1</sup>) 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 timeresolved 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 g min<sup>-1</sup> for the BDS and 14.9  $\pm$  0.6 g min<sup>-1</sup> 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 g min<sup>-1</sup>) and TSF tests (19.6  $\pm$  1.2 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 g  $min^{-1}$ ) was 23% higher than during BDS tests (5.5 ± 0.5 g min<sup>-1</sup>), 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  $CO_2$ , based on instantaneous emission factors calculated with eq 6 (i.e.,  $\mu g s^{-1}$ ) 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  $CO_2$  had been emitted. The  $CO_2$  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.

#### **Environmental Science & Technology**



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<sup>-1</sup>) markedly increased relative to the CO<sub>2</sub> emission rate prior to boiling (at about a quarter of the way into the test) and decreased relative to the CO<sub>2</sub> 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 CO<sub>2</sub> 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.<sup>30</sup> 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<sup>-1</sup>) 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<sup>-1</sup>) 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.<sup>31</sup>

**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<sup>-1</sup>) 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<sub>2.5</sub> emission factor (g kg<sup>-1</sup>) 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<sub>2.5</sub> emitted by the TSF. Figure 2a illustrates that the mass of PM<sub>2.5</sub> emitted during 45% of TSF tests (i.e., 9 out of 20) exceeded the mass of PM<sub>2.5</sub> emitted during all tests

Table 1	l. Summary	of Emissions of	of $CO_2$ ,	CO, PM <sub>2.5</sub>	, and BC by	Test Segment	for the BDS and TSF
---------	------------	-----------------	-------------	-----------------------	-------------	--------------	---------------------

				normalized avg emission rate <sup>b</sup>			normalized avg mass emissions <sup>c</sup>				
stove	test segment	avg duration (min)	normalized avg duration <sup>a</sup>	CO <sub>2</sub>	СО	PM <sub>2.5</sub>	BC	CO <sub>2</sub>	СО	PM <sub>2.5</sub>	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

"Average duration of test segment normalized by average test duration. <sup>b</sup>Segment average instantaneous emission rate  $(g s^{-1})$  normalized by the test average instantaneous emission rate. <sup>c</sup>Segment 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<sup>-1</sup>) during tests with the BDS was 115  $\pm$  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  $\pm$ 15% of the BC mass emitted by the TSF. BC emissions varied considerably from test to test and, compared to PM<sub>2.5</sub> emissions, there was greater overlap in distributions of BC emissions from the BDS and TSF (Figures 2a and b).

Given these results for PM<sub>2.5</sub> and BC, the emission ratio of BC/PM<sub>2.5</sub> was larger for the BDS than the TSF (Figure S12, SI). Consequently, the  $\mbox{MAE}_{\mbox{PM2.5}}$  (532 nm) of emitted by the BDS  $(4.6 \pm 0.3 \text{ m}^2 \text{ g}^{-1})$  was higher than that emitted by the TSF  $(3.1 \pm 0.4 \text{ m}^2 \text{ g}^{-1})$ . The MAE<sub>BC</sub> (532 nm) of BC was quite similar for both stoves (9.8  $\pm$  0.6  $m^2~g^{-1}$  for BDS and 9.0  $\pm$  0.7  $m^2 g^{-1}$  for TSF). Consistent with these results, the PM<sub>2.5</sub> emitted by the BDS had a lower SSA (Figure 2c and Table 2) and a lower AAE (Figure S14, SI) compared to the PM<sub>2.5</sub> emitted by the TSF. The lower SSA (average of  $0.36 \pm 0.02$  for BDS versus  $0.47 \pm 0.03$  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.<sup>27,32</sup>

**Effect of Wood Type.** Table S6 (SI) presents wood typespecific 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  $\pm$  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  $\pm$  11% of the PM<sub>2.5</sub> emitted by the TSF) indicate that the BDS design also led to a modest improvement in combustion efficiency. By comparison, Jetter et al.<sup>20</sup> 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  $PM_{25}$  (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 of al. and similar for the two stoves in the current study. Jetter et al. also found that the BDS had CO and PM2.5 emissions comparable to other wood-burning, natural-draft stoves.<sup>20</sup> 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.<sup>33</sup>

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.<sup>16,17,20</sup> However, these targets can be achieved through additional design modifications.<sup>34</sup> 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.<sup>20</sup> 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-towarming threshold of about 0.85,  $^{35,36}$  PM<sub>2.5</sub> 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  $MAE_{PM2.5}$  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  $CO_2$ 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 $(n = 2.1)$	TSF $(n = 2.0)$	BDS/TSF ratio
J	DD0 (n - 21)	101 (# - 20)	<i>DD0,</i> 101 Iudo
wood:			
moisture content	$0.09 \pm 0.01$	$0.09 \pm 0.01$	1.00
dry mass (g test <sup>-1</sup> )	368 ± 15	$564 \pm 32$	0.65
emission factors:			
$CO_2 (g kg^{-1})$	1767 ± 4	$1745 \pm 5$	1.01
$CO (g kg^{-1})$	$42 \pm 3$	56 ± 3	0.75
PM <sub>2.5</sub> (g kg <sup>-1</sup> )	$3.1 \pm 0.5$	$3.9 \pm 0.4$	0.79
BC (g kg <sup><math>-1</math></sup> )	$1.5 \pm 0.2$	$1.3 \pm 0.1$	1.15
BC/PM <sub>2.5</sub>	$0.48 \pm 0.03$	$0.35 \pm 0.04$	1.37
total mass emitted:			
$CO_2$ (g test <sup>-1</sup> )	$650 \pm 25$	985 ± 57	0.66
CO (g test <sup>-1</sup> )	16 ± 1	$32 \pm 2$	0.50
$PM_{2.5} (g \text{ test}^{-1})$	$1.2 \pm 0.2$	$2.3 \pm 0.3$	0.52
BC (g test <sup><math>-1</math></sup> )	$0.5 \pm 0.1$	$0.7 \pm 0.1$	0.71
emitted particle optic	al properties:		
$MAE_{PM2.5} (m^2 g^{-1})$	$4.6 \pm 0.3$	$3.1 \pm 0.4$	1.48
$MAE_{BC} (m^2 g^{-1})$	$9.8 \pm 0.6$	$9.0 \pm 0.7$	1.09
$MSE_{PM2.5} (m^2 g^{-1})$	$2.6 \pm 0.2$	$2.7 \pm 0.1$	0.96
SSA	$0.36 \pm 0.02$	$0.47 \pm 0.03$	0.77
1-SSA	$0.64 \pm 0.02$	$0.53 \pm 0.03$	1.21

dissemination.<sup>37,38</sup> While having the potential climate mitigation benefits of reduced  $CO_2$  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.<sup>3</sup> 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.<sup>3,7</sup> Given that BC is a very short-lived species relative to  $CO_2$  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<sub>2.5</sub> emission targets for the highest performance tier of biomass stoves.<sup>16,17</sup> For instance, reducing the average masses of BC (0.7 g, Table 2) and  $CO_2$ (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  $CO_2$  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 CO<sub>2</sub> 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

#### **Environmental Science & Technology**

are highly variable and that stove performance in the field may be quite different than it is in the laboratory.<sup>39-41</sup> These labfield 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.<sup>42</sup> 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.<sup>42</sup> 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.<sup>42</sup>

#### ASSOCIATED CONTENT

#### **S** 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/.

#### AUTHOR INFORMATION

#### Corresponding Author

\*Phone: (510) 486-5319. Fax: (510) 486-7303. E-mail: twkirchstetter@lbl.gov.

#### **Present Address**

<sup>§</sup>Olympic Region Clean Air Agency, Olympia, WA 98502

#### Notes

The authors declare no competing financial interest.

#### ACKNOWLEDGMENTS

This work was supported by the California Energy Commission under Contract No. 500-99-013. We gratefully acknowledge Douglas Sullivan and Jessica Granderson of Lawrence Berkeley National Laboratory for their support of this project, as well as the many students, interns, and researchers who, before us, contributed to the development of the Berkeley-Darfur Stove.

#### REFERENCES

(1) Smith, K. R.; Mehta, S.; Maeusezahl-Feuz, M. Indoor air pollution from household use of solid fuels. In *Comparative Quantification of Health Risks: Global and Regional Burden of Disease Due to Selected Major Risk Factor*; Ezzati, M., Rodgers, A. D., Lopez, A. D., Murray, C. L. J., Eds.; World Health Organization: Geneva, Switzerland, 2004.

(2) Lim, S. S.; Vos, T.; Flaxman, A. D.; Danaei, G.; et al. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. *Lancet* **2012**, 380, 2224–2260.

(3) Bond, T. C.; et al. Bounding the role of black carbon in the climate system: A scientific assessment. J. Geophys. Res. Atmos. 2013, 118, 5380-5552.

(4) Bond, T. C.; Streets, D. G.; Yarber, K. F.; Nelson, S. M.; Woo, J.; Klimont, Z. A technology-based global inventory of black and organic carbon emissions from combustion. *J. Geophys. Res.* **2004**, *109*, D14203.

(5) Menon, S.; Hansen, J.; Nazarenko, L.; Luo, Y. F. Climate effects of black carbon aerosols in China and India. *Science* **2002**, *297*, 2250–2253.

(6) Hansen, J.; Nazarenko, L. Soot climate forcing via snow and ice albedos. *Proc. Natl. Acad. Sci. U.S.A.* 2003, 101, 423-428.

(7) Ramanathan, V.; Carmichael, G. Global and regional climate changes due to black carbon. *Nat. Geosci.* **2008**, *1*, 221–227.

(8) Smith, K. R.; McCracken, J. P.; Weber, M. W.; Hubbard, A.; Jenny, A.; Thompson, L. M.; Balmes, J.; Diaz, A.; Arana, B.; Bruce, N. Effect of reduction in household air pollution on childhood pneumonia in Guatemala (RESPIRE): a randomized controlled trial. *Lancet* **2011**, *378*, 1717–1726.

(9) Amrose, S.; Kisch, G. T.; Kirubi, C.; Woo, J.; Gadgil, A. *Development and testing of the Berkeley Darfur stove*; Lawrence Berkeley National Laboratory: Berkeley, CA, 2008; http://www.escholarship. org/uc/item/3rf3x1df (accessed January 16, 2014).

(10) Masera, O.; Edwards, R.; Arnez, C. A.; Berrueta, V.; Johnson, M.; Bracho, L. R.; Riojas-Rodríguez, H.; Smith, K. R. Impact of Patsari improved cookstoves on indoor air quality in Michoacan, Mexico. *Energy Sustainable Dev.* **2007**, *11*, 45–56.

(11) Venkataraman, C.; Sagar, A. D.; Habib, G.; Lam, N.; Smith, K. R. The Indian National Initiative for Advanced Biomass Cookstoves: the benefits of clean combustion. *Energy Sustainable Dev.* **2010**, *14*, 63–72.

(12) MacCarty, N.; Ogle, D.; Still, D.; Bond, T.; Roden, C. A laboratory comparison of the global warming impact of five major types of biomass cooking stove. *Energy Sustainable Dev.* **2008**, *12*, 56–65.

(13) Ramanathan, V.; Balakrishnan, K. Project Surya: reduction of air pollution and global warming by cooking with renewable sources: a controlled and practical experiment in Rural India; a white paper. 2007; http://www.projectsurya.org/storage/Surya-WhitePaper.pdf (accessed January 16, 2014).

(14) Alliance mission and goals; http://www.cleancookstoves.org/the-alliance/ (accessed January 16, 2014).

(15) Biomass cookstoves technical meeting: summary report, Energy Efficiency and Renewable Energy Biomass Program; U.S. Department of Energy: Washington, DC, 2011; http://www1.eere.energy.gov/biomass/pdfs/cookstove\_meeting\_summary.pdf (accessed January 16, 2014).

(16) International Workshop Agreement 11:2012, Guidelines for Evaluating Cookstove Performance; International Organization for Standardization: Geneva, Switzerland, 2012; http://www.pciaonline. org/files/ISO-IWA-Cookstoves.pdf (accessed January 16, 2014).

(17) Mehta, S.; Chiang, R. *Measuring progress during phase I: Building on the IWA interim guidelines.* Global Alliance for Clean Cookstoves: Washington, DC, 2012; http://www.cleancookstoves.org/resources

#### **Environmental Science & Technology**

files/measuring-progress-during-phase-I.pdf (accessed January 16, 2014).

(18) Galitsky, C.; Gadgil, A.; Jacobs, M.; Lee, Y. Fuel Efficient Stoves for Darfur Camps of Internally Displaced Persons Report of Field Trip to North and South Darfur, Nov 16–Dec 17, 2005; Lawrence Berkeley National Laboratory: Berkeley, CA, 2006; p LBNL-59540

(19) Jetter, J. J.; Kariher, P. (2009) Solid-fuel household cook stoves: characterization of performance and emissions. *Biomass. Bioenerg.* 2009, 33, 294–305.

(20) Jetter, J.; Zhao, Y.; Smith, K. R.; Khan, B.; Yelverton, T.; DeCarlo, P.; Hays, M. D. Pollutant emissions and energy efficiency under controlled conditions for household biomass cookstoves and implications for metrics useful in setting international test standards. *Environ. Sci. Technol.* **2012**, *46*, 10827–10834.

(21) Arnott, W. P.; Moosmüller, H.; Rogers, C. F.; Jin, T.; Bruch, R. Photoacoustic spectrometer for measuring light absorption by aerosol: instrument description. *Atmos. Environ.* **1999**, *33*, 2845–2852.

(22) Penaloza, M. A. Deriving the basic cell-reciprocal integrating nephelometer equation and its use for calibration purposes: a comprehensive approach. *Meas. Sci. Technol.* **1999**, *10*, R1–R15.

(23) Arnott, W. P.; Moosmüller, H.; Walker, J. W. Nitrogen dioxide and kerosene-flame soot calibration of photoacoustic instruments for measurement of light absorption by aerosols. *Rev. Sci. Instrum.* 2000, 71, 4545–4552.

(24) Virkkula, A.; Ahlquist, N. C.; Covert, D. S.; Arnott, W. P.; Sheridan, P. J.; Quinn, P. K.; Coffman, D. J. Modification, calibration and field test of an instrument for measuring light absorption by particles. *Aerosol Sci. Technol.* **2005**, *39*, 68–83.

(25) Kirchstetter, T. W.; Novakov, T. Controlled generation of black carbon particles from a diffusion flame and applications in evaluating black carbon measurement methods. *Atmos. Environ.* **2007**, *41*, 1874–1888.

(26) Arnott, W. P.; Moosmüller, H.; Sheridan, P. J.; Ogren, J. A.; Raspet, R.; Slaton, W. V.; Hand, J. L.; Kreidenweis, S. M.; Collett, J. L., Jr. Photoacoustic and filter-based ambient aerosol light absorption measurements: Instrument comparisons and the role of relative humidity. J. Geophys. Res. 2003, 108, 4034.

(27) Kirchstetter, T. W.; Thatcher, T. L. Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation. *Atmos. Chem. Phys.* **2012**, *12*, 6067–6072.

(28) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O. Emission factors and real-time optical properties from traditional wood burning cookstoves. *Environ. Sci. Technol.* **2006**, *40*, 6750–6757.

(29) Gaur, S.; Reed, T. B. Thermal Data for Natural and Synthetic Fuels; Marcel Dekker: New York, 1998.

(30) McMeeking, G. R.; Kreidenweis, S. M.; Baker, S.; Carrico, C. M.; Chow, J. C.; Collett, J. L.; Hao, W. M.; Holden, A. S.; Kirchstetter, T. W.; Malm, W. C.; Moosmüller, H.; Sullivan, A. P.; Wold, C. E. Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory. *J. Geophys. Res.* **2009**, *114*, D19210.

(31) Andreae, M. O.; Merlet, P. Emission of trace gases and aerosols from biomass burning. *Global Biogeochem. Cy.* **2001**, *15*, 955–966.

(32) Andreae, M. O.; Gelenscér, A. Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols. *Atmos. Chem. Phys.* **2006**, *6*, 3131–3148.

(33) *Potential Energy* Website; http://www.potentialenergy.org (accessed January 16, 2014).

(34) Kshirsagar, M. P.; Kalamkar, V. R. A comprehensive review on biomass cookstoves and a systematic approach for modern cookstove design. . *Renew. Sust. Energ. Rev.* **2014**, *30*, 580–603.

(35) Houghton, J. T., Filho, L. G. M., Callander, B. A., Harris, N., Kattenberg, A., Maskell, K., Eds. Climate change: the science of climate change, contribution of working group I to the second assessment report of the Intergovernmental Panel on Climate Change; Cambridge University Press: New York, 2001.

(36) Hansen, J.; Sato, M.; Ruedy, R. Radiative forcing and climate response. J. Geophys. Res. **1997**, 102, 6831–6864.

(37) Akbar, S.; Ebinger, J.; Kleiman, G.; Oguah, S. Integration of shortlived climate pollutants in World Bank activities: a report prepared at the request of the G8; World Bank: Washington, DC, 2013; http://documents.worldbank.org/curated/en/2013/06/18119798/ integration-short-lived-climate-pollutants-world-bank-activities-reportprepared-request-g8 (accessed January 16, 2014).

(38) Johnson, M.; Edwards, R.; Ghilardi, A.; Berrueta, V.; Gillen, D.; Frenk, C. A.; Masera, O. Quantification of carbon savings from improved biomass cookstove projects. *Environ. Sci. Technol.* **2009**, *43*, 2456–2462.

(39) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O.; MacCarty, N.; Still, D. Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmos. Environ.* **2009**, *43*, 1170–1181.

(40) Chen, Y.; Roden, C. A.; Bond, T. C. Characterizing biofuel combustion with patterns of real-time emission data (PaRTED). *Environ. Sci. Technol.* **2012**, *46*, 6110–6117.

(41) MacCarty, N.; Still, D.; Ogle, D.; Drouin, T. Assessing cook stove performance: field and lab studies of three rocket stoves comparing the open fire and traditional stoves in Tamil Nadu, India on measures of time to cook, fuel use, total emissions, and indoor air pollution. Aprovecho Research Center, 2008; http://www.aprovecho. org/lab/pubs/arcpubs (accessed January 16, 2014).

(42) Wang, Y.; Sohn, M. D.; Wang, Y.; Lask, K. M.; Kirchstetter, T. W.; Gadgil, A. J. How many replicate tests are needed to test cookstove performance and emissions? – Three is not always adequate. *Energy Sustainable Dev.* **2014**, *20*, 21–29.

1 2	Emissions and Climate-Relevant Optical Properties of Pollutants Emitted from a Three- Stone Fire and the Berkeley-Darfur Stove Tested under Laboratory Conditions
3	
4	SUPPORTING INFORMATION
5	
6	Chelsea V. Preble <sup>1,2</sup> , Odelle L. Hadley <sup>2,a</sup> , Ashok J. Gadgil <sup>1,2</sup> , Thomas W. Kirchstetter <sup>1,2,*</sup>
7	
8 9	<sup>1</sup> Department of Civil and Environmental Engineering, University of California, Berkeley, California 94720-1710
10	
11 12	<sup>2</sup> Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720
13	
14	<sup>a</sup> Current address: Olympic Region Clean Air Agency, Olympia WA 98502
15 16 17 18 19 20	*Corresponding author e-mail: twkirchstetter@lbl.gov *Corresponding author phone number: (510) 486-5319 *Corresponding author fax number: (510) 486-7303
21	Number of pages: 22
22	Number of figures: 14
23	Number of tables: 7

# 25 INTRODUCTION

26 Berkeley-Darfur Stove (BDS). The BDS was developed at Lawrence Berkeley National 27 Laboratory (LBNL) and the University of California, Berkeley to aid internally displaced 28 populations from the Sudan civil war living in displacement camps in Darfur. While cooking 29 with the TSF. Darfuri women walk up to seven hours three to five times per week to collect a 30 sufficient amount of wood for cooking. Outside the relative safety of the camps, they are 31 vulnerable to acts of violence and sexual assault. To avoid this risk of attack during firewood 32 collection trips, a significant number of families now purchase firewood from vendors by trading 33 food rations for fuel (Galitsky et al., 2006).

34 The BDS was designed and tested to reduce the amount of wood needed for cooking 35 through increased thermal efficiency compared to cooking over a TSF – and thus reduce the risk 36 and/or cost associated with acquiring fuelwood in Darfur. The physical design of the BDS tested 37 in this study (version 14) is based on India's Tara Stove but modified for lower cost, longer life, 38 easier assembly, the cultural cooking requirements of Darfuri women, and the windy conditions 39 of the region (Amrose et al., 2008). As with the Tara Stove, the BDS contains the fire in a low-40 emissivity stainless steel firebox, thereby focusing the radiative heat of the fire to the pot. 41 Additionally, the BDS uses a particular pot and stove interface geometry to increase convective 42 heat transfer to the pot rather than allowing hot combustion gases to escape to the environment. 43 The fire in the BDS is maintained upon a raised grate to promote the supply of primary air to the 44 fire from below. The design also includes a tapered collar that allows for multiple pot sizes, 45 internal ridges for spacing between the stove and pot, nonaligned air openings between the outer 46 stove and inner fire box to reduce the effects of wind, and a small fire box opening to prevent the 47 user from overstuffing the stove with fuelwood (UCB, 2012). The BDS is constructed from a flat

**S**2

48 kit of sheet metal (see cookstoves.lbl.gov/Darfur.php) and can be purchased from Potential
49 Energy (http://www.potentialenergy.org/).

50 **Three-Stone Fire (TSF).** The TSF in this study was comprised of three large stones of similar 51 size that were placed into a triangular configuration, as shown in Figure S1. To better mimic the 52 thermal conductivity of the sandy ground that is typical of cooking in Darfur, as well as to 53 prevent heat loss to the metal sampling platform, fires were built upon a round ceramic stone 54 with a diameter of 22 cm. The round-bottomed Darfuri pot was balanced upon the three stones 55 that were configured around this ceramic base, such that the bottom of the pot was typically 10-56 13 cm above the base of the fire.

57

## 58 METHODS

59 Testing Facility and Measurements. All emission measurements were made at the cookstove 60 testing facility at Lawrence Berkeley National Laboratory. As shown in Figure S2, cooking was 61 carried out on a platform underneath a large plenum. A blower connected to the plenum via an 62 aluminum duct (diameter 15 cm) drew the smoke from cooking fires and room air into the 63 plenum. The smoke and room air mixed along the length of the duct, aided by stationary fan 64 blades. A calibrated airflow measurement system (The Energy Conservatory, Duct Blaster 65 System) was used to establish the relationship between the flow rate of air into the exhaust 66 plenum and the pressure and temperature at a fixed point inside the exhaust duct. The 67 instantaneous flow rate of air into the exhaust system (Q in Equation 6) during stove tests was 68 estimated from this calibration relationship and the duct pressure and temperature. 69 At a point prior to the blower, the ducted smoke was drawn via stainless steel tubing

70 (diameter 1.1 cm) through a sharp cut cyclone (BGI, model VSCCA) to remove particles with

71 diameters larger than 2.5 µm. Downstream of the cyclone, the sample flow was split using

**S**3

stainless steel wyes (BMI, models 1100, 1101, and 1102) and sampled with the instruments
listed in Table S1. Dry, particle-free air was mixed with the portion of the flow sampled by the
real-time particle sensing instruments. This provided further dilution of the PM concentration by
about a factor of 10 and reduced the relative humidity to well below 40%, such that the sampled
PM would not be coated with water (Seinfeld and Pandis, 2006).

The flow rates of each sample line and the secondary particle dilution rate illustrated in
Figure S2 were measured prior to each test with a primary standard air flow calibrator
(Sensidyne, Gilibrator-2). The measured flows were used to calculate filter-based BC and PM<sub>2.5</sub>
concentrations. The optical coefficients, BC concentrations, and PM<sub>2.5</sub> concentrations, measured
in diluted airflows, respectively with the PAS, aethalometer, and DustTrak, were corrected to
their values for undiluted airflow using the flow rates data.

83 In addition to the real-time (1 Hz) measurements of CO, CO<sub>2</sub>, BC, and PM<sub>2.5</sub>

84 concentrations and particle absorption and scattering coefficients, time-integrated concentrations

85 of BC and PM<sub>2.5</sub> were also periodically measured during cooking tests to develop calibration

86 equations specific to woodsmoke for the aethalometer and DustTrak, as described below.

88

87 Woodsmoke Specific Calibrations for BC and PM<sub>2.5</sub>. The BC concentrations reported by the

aethalometer were adjusted with Equation S1 to correct for a known sampling artifact that causes

89 the aethalometer's response to diminish as the filter darkens during sampling (Kirchstetter and

90 Novakov, 2007). Adjusted concentrations (BC<sub>corrected</sub>) are derived from unadjusted concentrations

91 (BC<sub>o</sub>) and the attenuation of light by the particle-laden filter (ATN), which the aethalometer

92 simultaneously reports. The applicability of this method was verified by comparing BC

93 concentrations measured with the aethalometer to absorption coefficients measured

simultaneously with the PAS. Both instruments respond to light absorbing particulate matter, but

**S**4

95 the *in-situ* PAS measurement is not biased by a filter. As shown in Figure S3, the ratio of 96 absorption coefficient to BC<sub>0</sub> increased by a factor of 2.5 as the aethalometer's filter progressed 97 from pristine (ATN = 0) to heavily loaded (ATN = 120). In contrast, the ratio of absorption 98 coefficient to BC<sub>corrected</sub> is approximately constant and independent of ATN. This comparison 99 validates the application of Equation S1 and illustrates the magnitude of the potential error, as 100 adjusted BC concentrations are up to 250% larger than unadjusted BC concentrations. 101 The rightmost term in Equation S1 further adjusts BC concentrations. The denominator, 102 flow (equal to 1.28), corrects for a difference in sampling flow rate measured by the aethalometer (1.0 L min<sup>-1</sup>) and using the air flow calibrator (1.28 L min<sup>-1</sup>). The numerator, *cal* (equal to 1.15), 103 104 accounts for the difference in BC concentrations measured by the aethalometer (after adjusting 105 for the sampling artifact, flow rate, and adjusting for secondary dilution) and those measured 106 using the thermal-optical analysis (TOA) method described by Hadley et al. (2008), which 107 distinguishes BC from other materials based on evolution temperature and spectral absorption 108 selectivity. The relationship between BC concentrations measured with the aethalometer and by 109 TOA is shown in Figure S4.

110

111 BC<sub>corrected</sub> = 
$$\left[\frac{BC_o}{0.88 \exp\left(\frac{ATN}{100}\right) + 0.12}\right] \left[\frac{cal}{flow}\right]$$
 (S1)

112

113 The  $PM_{2.5}$  concentrations measured with the DustTrak were adjusted using Equation S2. 114 Adjustment was necessary because this instrument is factory calibrated using a NIST certified 115 standard composed of soil from Arizona. Per the manufacturer's recommendation, alternate 116 calibrations should be applied when sampling different types of particulate matter. Equation S2 117 was developed by comparing  $PM_{2.5}$  concentrations measured with the DustTrak after adjusting

118	for secondary dilution ( $PM_o$ ) to those measured gravimetrically, as shown in Figure S5. The
119	mass of particles collected on Teflon filters was measured using a microbalance. To be
120	consistent with the dry air sampled by the DustTrak and accepted practice, the filters were
121	conditioned at a relative humidity $\leq 40\%$ prior to their weighing.
122	
123	$PM_{corrected} = 1.10(PM_{o})^{0.78} $ (S2)
124	
125	Optical Properties. The absorption Ångström exponent (AAE) is a measure of the variation in
126	particle light-absorption with wavelength (i.e., absorption selectivity). Absorption selectivity is
127	relatively weak for BC and strong for light absorbing organic particulate matter in wood smoke
128	(Kirchstetter and Thatcher, 2012). AAE is approximately 1 when BC is the primary light
129	absorber and larger than 1 when light-absorbing organic carbon is present, as it is in woodsmoke
130	(Hoffer et al., 2006; Andreae and Gelenscér, 2006). Absorption selectivity is often parameterized
131	using Equation S3, where $\lambda$ is the wavelength of light and k is a constant. Accordingly, AAE was
132	calculated from PSAP measurements of $b_{abs}$ at 467, 530, and 660 nm by performing a linear
133	regression of $ln(b_{abs})$ against $ln(\lambda)$ .
134	

135 
$$b_{abs}(\lambda) = k\lambda^{-AAE}$$
 (S3)

137 Test Protocol. The moisture content of each piece of wood was measured using a moisture 138 meter. The equivalent dry mass of each piece of wood  $(m_{dry})$  was calculated as:

$$140 \qquad m_{\rm dry} = \frac{m_{\rm wet}}{1 + f_{\rm m}} \tag{S4}$$

- 141 where  $m_{wet}$  is the mass of wood measured with the balance and  $f_m$  is the measured wood moisture
- 142 content.
- 143

# 144 **REFERENCES**

- Amrose, S.; Kisch, G.T.; Kirubi, C.; Woo, J.; Gadgil, A. Development and testing of the
- 146 Berkeley Darfur Stove. **2008**, Lawrence Berkeley National Laboratory: Berkeley, CA;
- 147 http://www.escholarship.org/uc/item/3rf3x1df.
- 148 Andreae, M.O.; Gelenscér, A. Black carbon or brown carbon? The nature of light-absorbing
- 149 carbonaceous aerosols. Atmos. Chem. Phys. 2006, 6, 3131–3148.
- 150 Galitsky, C.; Gadgil, A.; Jacobs, M.; Lee Y. Fuel efficient stoves for Darfur camps of internally
- 151 *displaced persons report of field trip to North and South Darfur, Nov. 16-Dec. 17, 2005.*
- 152 Hadley, O.L.; Corrigan, C.E.; Kirchstetter, T.W. Modified thermal-optical analysis using spectral
- absorption selectivity to distinguish black carbon from pyrolized organic carbon. *Environ. Sci.*
- 154 *Technol.* **2008**, 42, 8459–8464.
- 155 Hoffer, A.; Gelencsér, A.; Guyon, P.; Kiss, G.; Schmid, O.; Frank, G.P.; Artaxo, P.; Andreae,
- 156 M.O. Optical properties of humic-like substances (HULIS) in biomass-burning aerosols, Atmos.
- 157 Chem. Phys. 2006, 6, 3563-3570.
- 158 Kirchstetter, T.W.; Novakov, T. Controlled generation of black carbon particles from a diffusion
- 159 flame and applications in evaluating black carbon measurement methods. *Atmos. Environ.* 2007,
- 160 41, 1874-1888; DOI 10.1016/j.atmosenv.2006.10.067.
- 161 Seinfeld J.H., Pandis S.N.. Atmospheric Chemistry and Physics: From Air Pollution to Climate
- 162 Change; John Wiley and Sons: New York, **2006**.
- 163 UCB, University of California, Berkeley (2012) Cookstoves: Darfur Stove Design, Gadgil Lab;
- 164 http://gadgillab.berkeley.edu/research/energy/cookstoves/.
- 165



**Figure S1.** The three-stone fire (top picture) and the Berkeley-Darfur Stove, version 14 (bottom

171 picture) that were used in this study.





173 **Figure S2.** Schematic of the cookstove testing facility at Lawrence Berkeley National

174 Laboratory used in this study.



176 **Figure S3.** Data from a representative cooking test illustrating that aethalometer BC

177 concentrations ( $BC_0$ ) are erroneously dependent on the amount of BC collected on its filter 178 (attenuation) and varifying that adjusted concentrations ( $BC_0$ ) are not (also see Equation

178 (attenuation) and verifying that adjusted concentrations (BC<sub>corrected</sub>) are not (also see Equation
 179 S1).



Figure S4. Relationship between BC concentrations measured with the aethalometer and bythermal-optical analysis of quartz filters.



186

Figure S5. Relationship between PM<sub>2.5</sub> concentrations measured with the DustTrak and by
 gravimetric analysis of Teflon filters.



187 Figure S6. Water temperature during BDS (solid red lines) and TSF (solid black lines) cooking

tests, representative heating rates (dashed lines), and ranges of time required to boil (shadedregions).



190

191 **Figure S7.** Distribution of dry wood consumption (the legend shows average  $\pm$  95% confidence 192 interval) when cooking with the BDS and TSF.





194 **Figure S8.** Distribution of test durations when cooking with the BDS and TSF.





Figure S9. Distribution of CO fuel-based emission factor (left) and mass of CO emitted (right) incooking tests with the BDS and TSF.



199 **Figure S10.** Distribution of  $PM_{2.5}$  fuel-based emission factor (left) and mass of  $PM_{2.5}$  emitted 200 (right) in cooking tests with the BDS and TSF.



201

Figure S11. Distribution of BC fuel-based emission factor (left) and mass of BC emitted (right)
 in cooking tests with the BDS and TSF.



205 **Figure S12.** Distribution of BC/PM<sub>2.5</sub> emission ratio in cooking tests with the BDS and TSF.



Figure S13. Distribution of  $PM_{2.5}$  mass absorption efficiency (left) and mass scattering efficiency (right) in tests with the BDS and TSF.



Figure S14. Distribution of  $PM_{2.5}$  extinction-weighted single scattering albedo (left) and absorption Angstrom exponent (right) in tests with the BDS and TSF.

 Table S1. Instrumentation used to quantify pollutant emissions.

	Parameter	Instrument	Primary or Secondary Products		
	CO and CO <sub>2</sub> concentrations	NDIR analyzer (CAI 600 series)	Combustion efficiency; fuel-based pollutant emission factors (g kg <sup>-1</sup> )		
	PM <sub>2.5</sub> concentration	DustTrak (TSI 8534)			
ne-Resolved (1 Hz)	BC concentration	Aethalometer (Magee Scientific AE16)	BC and PM <sub>2.5</sub> fuel-based emission factors (g kg <sup>-1</sup> ); particle mass		
	Absorption and scattering coefficients	Photoacoustic absorption, reciprocal nephelometry (custom)	particle single scattering albedo		
Tin	Absorption coefficient at 3- wavelengths	Particle-soot absorption photometer (Radiance Research)	Absorption spectral selectivity (particle color)		
	Water temperature	Type K thermocouple and digital thermometer	Water heating rate; test duration		
	Duct pressure and temperature	Pressure sensor and type K thermocouple	Dilution flow rate		
ted	PM <sub>2.5</sub> mass	Teflon filter; microbalance (Sartorius SE2-F)	Calibration of DustTrak		
ne-Integra	BC mass	Quartz filter; thermal- optical analysis	Calibration of Aethalometer		
Tin	Wood mass	Balance (Sartorius ED5201) and moisture meter (Delmhorst J-2000)	Dry wood addition rate		

214

216 **Table S2.** Test duration, mass and moisture content of wood combusted, and average pollutant

217 concentrations and optical properties measured during BDS tests.

Test	Duration (min)	Dry Wood Combusted (g)	Wood Moisture (%)	ΔCO <sub>2</sub> (ppm)	CO (ppm)	PM <sub>2.5</sub> (mg m <sup>-3</sup> )	BC (mg m <sup>-3</sup> )	Abs* (Mm <sup>-1</sup> )	Scat* (Mm <sup>-1</sup> )
01	31.4	351	7.1	477	74	5.1	3.1	29165	14142
02	31.2	322	7.4	484	60	4.1	2.4	25013	10790
03	29.9	438	11.2	464	97	7.0	3.1	24096	18344
04	32.6	390	10.2	520 61 5.8 2.3 20517		20517	11466		
05	34.2	429	10.3	491	73	5.9	2.5	24552	13741
06	33.3	421	10.7	482	84	6.4	3.3	26082	16750
07	29.7	375	8.6	534	69	6.5	3.5	36534	20105
08	29.5	338	7.2	542	58	4.8	2.4	28780	12435
09	33.4	413	9.9	473	473 73 6.0		2.3	25311	13531
10	30.5	375	9.2	518	89	6.9	2.5	25509	15003
11	29.1	326	9.1	534	71	4.5	1.4	19624	9472
12	27.9	337	8.9	549	76	4.4	2.6	22658	11424
13	30.6	352	9.2	584	67	3.0	1.6	15397	6337
14	28.4	359	9.4	536	78	4.9	2.6	23251	11375
15	30.3	352	9.5	580	71	4.1	2.1	17916	9779
16	28.3	353	10.5	597	72	5.0	2.3	21332	11275
17	26.9	333	8.0	593	117	9.8	4.6	43280	28240
18	30.0	363	8.1	551	81	11.8	5.7	62651	40342
19	33.2	378	7.8	573	77	10.9	4.7	51635	31790
20	28.3	373	8.0	575	112	12.8	5.0	44936	39400
21	27.4	351	7.8	628	101	12.4	6.2	62961	42111
avg ± 95% CI	30.3 ± 0.9	368 ± 15	9.0 ± 1.0	537 ± 20	$79\pm7$	6.8 ± 1.3	3.2 ± 0.6	31010 ± 5989	18469 ± 4716

218

219 \* Particle absorption (Abs) and scattering (Scat) coefficients were measured at 532 nm.

220 **Table S3.** Test duration, mass and moisture content of wood combusted, and average pollutant

221 concentrations and optical properties measured during TSF tests.

Test	Duration (min)	Dry Wood Combusted (g)	Wood Moisture (%)	ΔCO <sub>2</sub> (ppm)	CO (ppm)	PM <sub>2.5</sub> (mg m <sup>-3</sup> )	BC (mg m <sup>-3</sup> )	Abs* (Mm <sup>-1</sup> )	Scat* (Mm <sup>-1</sup> )
01	40.0	540	7.0	455	105	8.0	3.5	31141	23811
02	42.1	505	7.2	480	107	6.1	2.8	23715	16418
03	48.1	624	11.3	471	132	12.0	2.8	16704	41539
04	48.2	655	9.5	484	92	9.3	2.5	20162	24894
05	45.7	633	9.6	500	108	9.5	2.8	22159	23364
06	46.5	522	9.7	529	79	7.6	2.1	18170	21065
07	34.7	496	7.0	544	114	7.7	3.2	30877	20156
08	50.4	737	10.8	471	97	9.2	2.4	25395	24264
09	35.2	558	9.0	511	116	11.3	3.0	25180	28612
10	33.3	458	9.1	558	112	7.4	1.9	19418	15541
11	42.6	538	9.3	476	89	6.3	2.2	17873	15850
12	34.8	482	9.3	572	108	5.5	2.2	18135	13425
13	42.4	511	9.0	546	96	5.3	2.1	16226	11822
14	40.3	568	9.9	536	100	6.6	2.7	21864	17204
15	37.3	500	10.8	568	97	5.0	1.9	18844	12062
16	35.5	538	10.9	553	139	8.2	2.8	22771	20921
17	34.6	494	7.8	589	102	9.9	3.0	43155	27338
18	50.3	671	7.9	496	104	13.2	3.8	36571	39460
19	39.0	640	7.6	551	99	9.8	5.7	54320	28298
20	39.8	615	7.9	581	121	12.2	3.6	33866	35675
avg ± 95% CI	41.0 ± 2.4	564 ± 32	9.0 ± 1.0	524 ± 18	106 ± 6	8.5 ± 1.0	$2.9 \pm 0.4$	25827 ± 4241	23086 ± 3665

222

223 \* Particle absorption (Abs) and scattering (Scat) coefficients were measured at 532 nm.

	Emission Factor			Tota	l Mass En	nitted	MAE	MAE	MSE	
Test		(g kg <sup>-1</sup> )			<b>(g</b> )		$(PM_{2.5})^*$	( <b>BC</b> ) <sup>*</sup>	$(PM_{2.5})^*$	$\mathbf{SSA}^*$
	CO	PM <sub>2.5</sub>	BC	СО	<b>PM</b> <sub>2.5</sub>	BC	$(\mathbf{m}^2 \mathbf{g}^{-1})$	$(\mathbf{m}^2 \mathbf{g}^{-1})$	$(m^2 g^{-1})$	
01	40	2.5	1.5	14	0.9	0.52	5.7	9.5	2.8	0.33
02	35	2.1	1.2	11	0.7	0.39	6.0	10.4	2.6	0.30
03	46	2.9	1.3	20	0 1.3 0.57 3.4		7.7	2.6	0.43	
04	36	3.0	1.2	14	1.2	0.46	3.5	9.0	2.0	0.36
05	40	2.8	1.2	17	1.2	0.51	4.2	9.8	2.3	0.36
06	44	2.9	1.5	19	1.2	0.64	4.1	7.9	2.6	0.39
07	38	3.1	1.7	14	1.2	0.63	5.6	10.4	3.1	0.35
08	33	2.4	1.2	11	0.8	0.41	6.0	11.8	2.6	0.30
09	39	2.8	1.1	16	1.2	0.45	4.2	11.0	2.2	0.35
10	50	3.4	1.2	19	1.3	0.46	3.7	10.4	2.2	0.37
11	41	2.3	0.7	13	0.7	0.24	4.4	13.6	2.1	0.33
12	42	2.1	1.3	14	0.7	0.42	5.2	8.6	2.6	0.34
13	39	1.5	0.8	14	0.5	0.30	5.1	9.3	2.1	0.29
14	41	2.2	1.2	15	0.8	0.43	4.8	8.9	2.3	0.33
15	44	2.2	1.2	15	0.8	0.41	4.4	8.4	2.4	0.35
16	40	2.5	1.1	14	0.9	0.40	4.2	9.1	2.2	0.35
17	56	4.1	1.9	19	1.4	0.64	4.4	9.4	2.9	0.39
18	41	5.2	2.5	15	1.9	0.91	5.3	11.0	3.4	0.39
19	42	5.2	2.3	16	2.0	0.86	4.7	10.9	2.9	0.38
20	53	5.3	2.1	20	2.0	0.77	3.5	9.0	3.1	0.47
21	50	5.4	2.7	18	1.9	0.94	5.1	10.2	3.4	0.40
avg ± 95% CI	42 ± 3	3.1 ± 0.5	1.5 ± 0.2	16 ± 1	1.2 ± 0.2	0.50 ± 0.09	4.6 ± 0.3	9.8 ± 0.6	2.6 ± 0.2	0.36 ± 0.02

225 **Table S4.** Emissions and optical properties of pollutants during BDS tests.

227 \* Particulate matter mass absorption and scattering efficiencies (MAE and MSE) and single

scattering albedo (SSA) were measured at 532 nm. SSA values reported here are weighted by

total extinction.

	Emission Factor			Tota	l Mass En	nitted	MAE	MAE	MSE	
Test		(g kg <sup>-1</sup> )			<b>(g</b> )		$(PM_{2.5})^*$	<b>(BC)</b> <sup>*</sup>	$(PM_{2.5})^*$	$\mathbf{SSA}^*$
	CO	PM <sub>2.5</sub>	BC	СО	<b>PM</b> <sub>2.5</sub>	BC	$(\mathbf{m}^2  \mathbf{g}^{-1})$	$(\mathbf{m}^2 \mathbf{g}^{-1})$	$(m^2 g^{-1})$	
01	56	3.7	1.6	30	2.0	0.87	3.9	9.0	3.0	0.43
02	59	3.0	1.4	30	1.5	0.69	3.9	8.4	2.7	0.41
03	76	6.0	1.4	47	3.7	0.88	1.4	5.9	3.5	0.71
04	52	4.6	1.2	34	34 3.0		2.2	8.0	2.7	0.55
05	59	4.5	1.3	37	2.9	0.84	2.3	7.9	2.5	0.51
06	55	4.6	1.3	29	2.4	0.68	2.4	8.5	2.8	0.54
07	56	3.3	1.3	28	28 1.6 0.67		4.0	9.8	2.6	0.39
08	51	4.2	1.1	37	37 3.1 0.82		2.8	10.4	2.6	0.49
09	60	5.1	1.4	33	2.8	0.75	2.2	8.4	2.5	0.53
10	58	3.3	0.8	27	1.5	0.39	2.6	10.5	2.1	0.44
11	52	3.2	1.1	28	1.7	0.60	2.8	8.2	2.5	0.47
12	58	2.6	1.0	28	1.2	0.51	3.3	8.1	2.4	0.43
13	57	2.7	1.1	29	1.4	0.55	3.1	7.8	2.2	0.42
14	56	3.2	1.3	32	1.8	0.75	3.3	8.0	2.6	0.44
15	55	2.5	0.9	27	1.2	0.46	3.8	10.0	2.4	0.39
16	65	3.3	1.2	35	1.8	0.62	2.8	8.1	2.6	0.48
17	49	4.1	1.2	24	2.0	0.62	4.4	14.4	2.8	0.39
18	52	5.8	1.7	35	3.9	1.12	2.8	9.6	3.0	0.52
19	41	3.5	2.1	26	2.3	1.32	5.5	9.5	2.9	0.34
20	55	4.8	1.4	34	3.0	0.88	2.8	9.4	2.9	0.51
avg ± 95% CI	56 ± 3	3.9 ± 0.4	1.3 ± 0.1	32 ± 2	2.3 ± 0.3	0.70 ± 0.09	3.1 ± 0.4	$9.0 \pm 0.7$	$2.7 \pm 0.1$	0.47 ± 0.03

230 **Table S5.** Emissions and optical properties of pollutants during TSF tests.

232 \* Particulate matter mass absorption and scattering efficiencies (MAE and MSE) and single

233 scattering albedo (SSA) were measured at 532 nm. SSA values reported here are weighted by

total extinction.

Table S6. Wood consumed, fuel-based pollutant emission factors, total mass of pollutants emitted, and particle optical properties (at 532
 nm), averaged over all tests and segregated by wood type, for all BDS and TSF tests conducted.

			BD	S			TS	F			BDS / TS	F Ratio	
Aver: W	age Values by /ood Type	Hard Oak (n = 6)	Mixed Fir/Pine (n = 9)	<b>Pine</b> <b>Only</b> (n = 6)	All Tests (n = 21)	Hard Oak (n = 5)	Mixed Fir/Pine (n = 11)	Pine Only (n = 4)	All Tests (n = 20)	Hard Oak	Mixed Fir/Pine	Pine Only	All Tests
poc	Moisture Content	0.10	0.09	0.08	0.09	0.10	0.09	0.08	0.09	1.01	0.96	1.07	1.00
M	Dry Mass (g test <sup>-1</sup> )	400	353	358	368	621	524	605	564	0.64	0.67	0.59	0.65
	CO <sub>2</sub> (g kg <sup>-1</sup> )	1769	1770	1759	1767	1747	1741	1756	1745	1.01	1.02	1.00	1.01
actor	CO (g kg <sup>-1</sup> )	41	40	47	42	55	59	49	56	0.75	0.68	0.96	0.75
sion F	PM <sub>2.5</sub> (g kg <sup>-1</sup> )	3.0	2.2	4.6	3.1	4.6	3.3	4.6	3.9	0.65	0.67	1.01	0.79
Emiss	BC (g kg <sup>-1</sup> )	1.3	1.2	2.1	1.5	1.3	1.2	1.6	1.3	1.03	0.96	1.31	1.15
	BC/PM <sub>2.5</sub>	0.44	0.52	0.46	0.48	0.28	0.37	0.37	0.35	1.58	1.40	1.24	1.37
	CO <sub>2</sub> (g test <sup>-1</sup> )	708	624	631	650	1085	911	1062	985	0.65	0.68	0.59	0.66
Mass tted	CO (g test <sup>-1</sup> )	16	14	17	16	34	31	30	32	0.48	0.46	0.56	0.50
Total Emi	PM <sub>2.5</sub> (g test <sup>-1</sup> )	1.2	0.8	1.7	1.2	2.8	1.8	2.8	2.3	0.42	0.45	0.59	0.52
	BC (g test <sup>-1</sup> )	0.5	0.4	0.8	0.5	0.8	0.6	1.0	0.7	067	0.64	0.76	0.71
ties	$\frac{MAE(PM_{2.5})}{(m^2 g^{-1})}$	4.2	5.0	4.5	4.6	2.4	3.2	3.9	3.1	1.78	1.58	1.18	1.48
roper	MAE(BC) (m <sup>2</sup> g <sup>-1</sup> )	9.8	9.8	9.9	9.8	8.6	7.3	10.7	9.0	1.13	1.34	0.93	1.09
tical P	$\frac{MSE(PM_{2.5})}{(m^2 g^{-1})}$	2.4	2.5	3.0	2.6	2.6	2.6	2.9	2.7	0.92	0.94	1.03	0.96
5 Opti	SSA	0.36	0.33	0.40	0.36	0.52	0.46	0.44	0.47	0.69	0.73	0.90	0.77
PM <sub>2</sub>	1 - SSA	0.64	0.67	0.60	0.64	0.48	0.54	0.56	0.53	1.34	1.23	1.08	1.21

**Table S7.** A comparison of the BDS and TSF time to boil, fuel burn/feed rates, and pollutant

emission factors (g kg<sup>-1</sup> on a dry fuel basis) reported by Jetter et al. (2012), as noted in their SI

240 (Figures S9, S14, S24, and S32), and found in the current study for the pre-boil and post-boil

241 phases of the similar constantly attended cooking tests. The total dry wood combusted and total

emitted CO and  $PM_{2.5}$  that are summarized below for Jetter et al. were calculated from the times

to boil and simmer and the pre- and post-boil fuel burn rates and emission factors.

Stove		<b>Jetter et al.</b> (2012) <sup>1</sup>			This study <sup>2</sup>		
Performance Parameter by Phase of Test		BDS	TSF	BDS/TSF Ratio	BDS	TSF	BDS/TSF Ratio
Time to Boil (min)		37.8	29.6	1.28	15.3	26.0	0.59
Fuel Burn/Feed	Pre- Boil	9.0	27.9	0.32	20.1	19.6	1.03
Rate <sup>3</sup> (g min <sup>-1</sup> )	Post- Boil	7.4	15.9	0.47	5.5	7.1	0.77
CO Emission	Pre- Boil	33	30	1.10	39	51	0.76
Factor (g kg <sup>-1</sup> )	Post- Boil	51	63	0.81	46	67	0.69
PM <sub>2.5</sub> Emission	Pre- Boil	1.5	1.8	0.83	3.8	4.2	0.90
Factor (g kg <sup>-1</sup> )	Post- Boil	2.3	2.7	0.85	2.2	3.3	0.67
Total Dry Wood Combusted <sup>4</sup> (g)		562	1303	0.43	368	564	0.65
Total CO Emitted (g)		22	55	0.40	16	32	0.50
Total PM <sub>2.5</sub> Emitted (g)		1.0	2.8	0.36	1.2	2.3	0.52

244

<sup>1</sup>Cold-start, high-power boil of 5 L of water followed by 30-minute low-power simmer (entire
test without a lid on the pot).

<sup>2</sup>Cold-start, higher power boil of 2.5 L of water followed by 15-min lower power simmer (entire
test with a lid on the pot).

<sup>3</sup>Jetter et al. report fuel burn rates whereas this study reports fuel feed rates.

<sup>4</sup>Total dry wood combusted (g) was calculated from Jetter et al.'s average pre- and post-boil fuel

burn rates and times to boil and simmer.