

## Evaluating the Performance of Household Liquefied Petroleum Gas Cookstoves

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### Supporting Information

**ABSTRACT:** Liquefied petroleum gas (LPG) cookstoves are considered to be an important solution for mitigating household air pollution; however, their performance has rarely been evaluated. To fill the data and knowledge gaps in this important area, 89 laboratory tests were conducted to quantify efficiencies and pollutant emissions from five commercially available household LPG stoves under different burning conditions. The mean thermal efficiency ( $\pm$ standard deviation) for the tested LPG cookstoves was  $51 \pm 6\%$ , meeting guidelines for the highest tier level (Tier 4) under the International Organization for Standardization, International Workshop Agreement 11. Emission factors of  $\text{CO}_2$ , CO, THC,  $\text{CH}_4$ , and  $\text{NO}_x$  on the basis of useful energy delivered ( $\text{MJ}_d$ ) were  $142 \pm 17$ ,  $0.77 \pm 0.55$ ,  $130 \pm 196$ ,  $5.6 \pm 8.2$ , and  $46 \pm 9$   $\text{mg}/\text{MJ}_d$ , respectively. Approximately 90% of the  $\text{PM}_{2.5}$  data were below the detection limit, corresponding to an emission rate below  $0.11$   $\text{mg}/\text{min}$ . For those data above the detection limit, the average emission factor was  $2.4 \pm 1.6$   $\text{mg}/\text{MJ}_d$ , with a mean emission rate of  $0.20 \pm 0.16$   $\text{mg}/\text{min}$ . Under the specified gas pressure (2.8 kPa), but with the burner control set to minimum air flow rate, less complete combustion resulted in a visually yellow flame, and CO,  $\text{PM}_{2.5}$ , EC, and BC emissions all increased. LPG cookstoves met guidelines for Tier 4 for both CO and  $\text{PM}_{2.5}$  emissions and mostly met the World Health Organization Emission Rate Targets set to protect human health.



### INTRODUCTION

Globally, nearly three billion people use solid fuels such as coal, charcoal, biomass, and dung for daily cooking and heating.<sup>1</sup> Solid fuels are typically burned in open fires or rudimentary stoves, resulting in fuel overconsumption and deleterious emission products due to incomplete combustion. Residential solid fuel combustion has been identified as a major source of air pollutants that affect human health and global climate, including CO (carbon monoxide),  $\text{PM}_{2.5}$  (particulate matter with an aerodynamic diameter  $\leq 2.5$   $\mu\text{m}$ ), and BC (black carbon).<sup>2–4</sup> According to the Global Burden of Disease study,<sup>5</sup> household air pollution is the top environmental risk factor, responsible for  $\sim 2.9$  million premature deaths and 81 million disability-adjusted life years lost in 2013. Additionally, residential fuel combustion contributed to  $\sim 30\%$  of 3.3 million premature deaths linked to outdoor air pollution.<sup>6</sup>

International and national efforts are currently directed toward deployment of clean fuels and cookstoves to reduce air

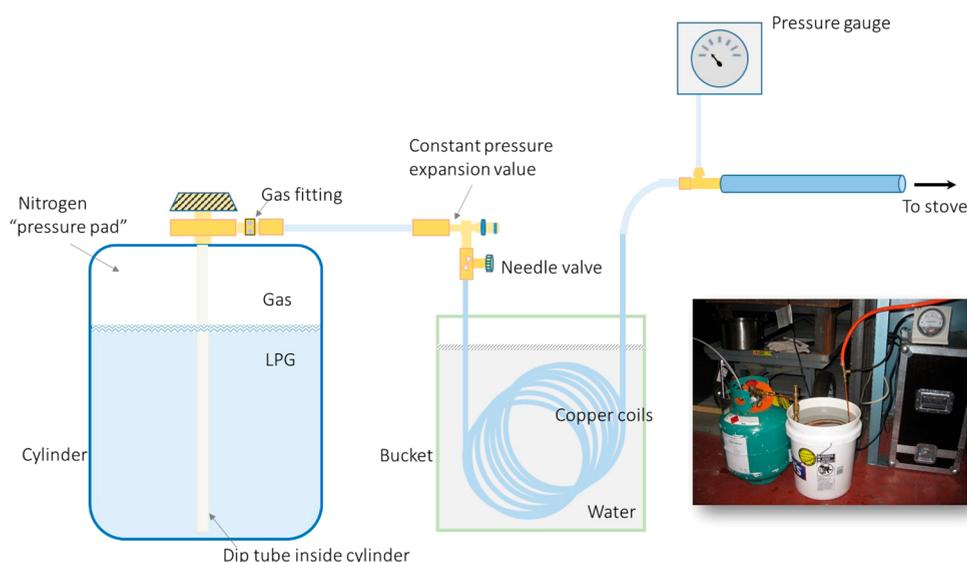
pollutant emissions from the residential sector and, consequently, to improve air quality, to protect human health, and to address climate change.<sup>7,8</sup> LPG (liquefied petroleum gas) fuel is considered to be among the most important fuels for achieving clean cooking.<sup>9</sup> Many countries are actively developing their national LPG intervention programs. For example, the Indonesian program converted over 50 million households cooking with kerosene to LPG within five years from 2007.<sup>10</sup> In 2014, the Ghana Ministry of Energy established a program to deploy LPG in rural homes by the provision of stoves and the optimization of supply networks,<sup>11</sup> and in 2016, India launched the PMUY (Pradhan Mantri Ujjwala Yojana)

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**Figure 1.** Schematic of the gas delivery system.

campaign to provide free connections to LPG cylinders to “Below Poverty Line” homes.<sup>12</sup>

LPG is produced with different compositions depending on economics, regional norms, and climate. It is typically a mixture of propane and butane but may contain low concentrations of other hydrocarbons. Olefins and other contaminant gases can be present as well, with a somewhat higher likelihood in LPG from oil refineries compared to LPG coproduced from natural gas production. A greater percentage of propane is typically used in cold climates due to its higher vapor pressure.

Few studies have reported efficiencies and air pollutant emissions from LPG cookstoves.<sup>13–18</sup> Zhang, Smith, and co-workers<sup>13,14</sup> quantified thermal efficiency and air pollutant emissions for one conventional-burner LPG stove and one infrared-head LPG stove in China (test replicate sample size,  $n$ , was 3 for each stove). The LPG tested was a mixture of 19% butane, 27% propane, 43% butene, and 11% other hydrocarbons. The infrared-head is a circular device attached around the burner under the pot to convert a part of heat released from the burner into infrared radiation that heats the pot bottom. Smith et al.<sup>15</sup> reported the efficiency and pollutant emissions from LPG (80% butane and 20% propane) burning in a household LPG stove ( $n = 3$ ) in India. Habib et al.<sup>16</sup> investigated  $PM_{2.5}$  and its chemical and optical properties from LPG burning ( $n = 1$ ) and compared with biomass burning in a mud stove in India. MacCarty et al.<sup>17</sup> reported  $CO_2$ , CO, and  $PM_{2.5}$  emissions from the burning of propane in a single-burner mass-produced camping stove ( $n = 1$ ).

These previous studies provided important novel emissions data for LPG cookstoves, but the studies had limited sample size and compositional differences in the fuel, and results were highly variable. For example,  $PM_{2.5}$  emissions ranged from  $0.54 \pm 0.24$  mg/MJ<sub>d</sub> (mass per useful energy delivered) to  $25 \pm 43$  mg/MJ<sub>d</sub>.<sup>13–15</sup> The influence of factors such as the stove power level, burner air control, and stove deterioration on emissions have not yet been investigated. Further understanding of LPG cookstove performance is required owing to high variability in air pollutant emissions during the LPG burning process. Moreover, some previous studies found comparable, or even occasionally higher, indoor levels of CO,  $NO_x$  (nitrogen oxides, including NO and  $NO_2$ ), and ultrafine particles in some homes

using LPG for cooking compared to those measured in homes using biomass or coal,<sup>19–21</sup> likely due to other sources of emissions.

This study aims to investigate efficiencies and air pollutant emission factors from LPG cookstoves under a variety of conditions. Five different household LPG stoves were tested, and the influence of different gas compositions, stove power levels, air control adjustments, and burner condition were examined. Knowledge gained from emission studies can contribute to a better understanding of the characteristics of LPG cookstove emissions, and may increase confidence in the effectiveness of LPG stove interventions.<sup>10–12,22</sup>

## METHODS

### Cookstove Test Facility and Emission Measurements.

The U.S. EPA CTF (Cookstove Test Facility) located in Research Triangle Park, NC, is designed for testing cookstove thermal efficiency and air pollutant emissions of a wide variety of fuels and stoves with or without chimneys. Results reported by the EPA and other testing facilities around the world are made available through publications and through the Global Alliance for Clean Cookstoves – Clean Cooking Catalog.<sup>23</sup> Results are comparable using the ISO IWA-11 (International Organization for Standardization, International Workshop Agreement) tier rating system.<sup>24</sup>

More detailed information about the CTF can be found elsewhere.<sup>25</sup> Briefly, emissions are collected and measured with a system consisting of a stainless steel hood connected to a dilution tunnel. Negative pressure is maintained throughout the entire system. An induced-draft blower provides dilution air and hood air flows. Volumetric flow of emissions and dilution air in the tunnel is nearly constant at  $\sim 4.3$  m<sup>3</sup>/min, and the dilution ratio varies with the output of emissions from the cookstove. Gaseous CO, CO<sub>2</sub>, THC (total hydrocarbons, based on propane), and CH<sub>4</sub> are continuously measured in the dilution tunnel using nondispersive infrared and flame ionization detector analyzers (Models 600, 600-HFID, and 600M-HFID, California Analytical, Orange, CA).  $NO_x$  emissions are measured in real time with a chemiluminescence  $NO_x$  analyzer (Model 200EH, Teledyne, San Diego, CA). Gas analyzers are calibrated and checked for zero and span at the



**Figure 2.** Pictures of five LPG burners tested in the present study. Stove A: Aodian stove from rural China. Stove B: AOSD stove from rural China. Stove C: Mikachi stove from a local market in Uganda. Stove D: Solgas stove from Peru. and Stove E: Simcook stove from a rural home in Cameroon.

start and end of each test day. Filter-based  $PM_{2.5}$  is sampled isokinetically from the dilution tunnel on a PTFE (polytetrafluoroethylene) membrane filter positioned downstream from a  $PM_{2.5}$  cyclone (University Research Glassware, Chapel Hill, NC) and measured gravimetrically using a microbalance with a readability of  $1 \mu\text{g}$  (MCS, Sartorius, Germany). To analyze carbon fractions in  $PM_{2.5}$ , particles are also sampled on a pretreated ( $550 \text{ }^\circ\text{C}$ , 12 h) quartz-fiber filter positioned downstream from another parallel cyclone. A second quartz filter is placed downstream of the PTFE filter to estimate the positive artifact due to gas-phase adsorption of semivolatile organics.<sup>26,27</sup> EC (elemental carbon) and OC (organic carbon) are quantified using a thermal-optical analyzer (Model 4L, Sunset Laboratory, Forest Grove, OR) following a modified National Institute for Occupational Safety and Health (NIOSH) Method 5040 protocol.<sup>28</sup> BC in emissions is measured optically in real time with a microAeth Model AE51 (Aethlabs, San Francisco, CA) sampling from a system that provides additional filtered dilution air. Emissions tests are conducted in a laboratory environment with temperature-controlled filtered air. The ambient temperature during the test period was  $21\text{--}23 \text{ }^\circ\text{C}$ .

As a typical practice for household LPG cookstoves, compressed (liquefied) gas is supplied in a cylinder, and gas is delivered from the headspace of the cylinder. The composition of a gas mixture changes as gas exits the cylinder due to the different vapor pressures of the component gases. This is a potential problem for determining the efficiency of a gas-fueled stove because the caloric values of gas fuels may vary, depending on the gas composition. Therefore, a gas delivery system was developed, as shown in Figure 1, to deliver gas mixtures with a constant composition. Each certified gas

mixture was obtained in a cylinder with a dip tube—the fuel was taken from the cylinder as a liquid (not as a gas). A nitrogen “pressure pad” filled the gas headspace above the liquid. The liquid fuel mixture did not change composition as the fuel was depleted in the cylinder. A manually adjusted needle valve downstream from a constant-pressure expansion valve was used to fine-tune the gas delivery pressure. When the liquid flashed to vapor, heat was absorbed, and the gas line became cold. The gas flowed through copper tubing coils submerged in water to deliver the gas at nearly ambient temperature. The expansion valve and needle valve were adjusted to maintain the line pressure as specified by the stove manufacturers ( $2.8 \text{ kPa}$ ). Near the end of each test phase, the gas valve was turned off at the cylinder to allow remaining liquid fuel (in the short line between cylinder and expansion valve) to flash to gas before the end of the phase.

**Stoves Tested.** Five different LPG stoves were tested (Figure 2) and are described below. Detailed photos (top, bottom, and the burner) are shown in Figure S1. Stoves A and B were manufactured in China and obtained in a local market near Beijing. Stove C was manufactured in Japan and obtained in a local market in Kampala, Uganda. Stove D was disseminated in Peru by a project involving Solgas Repsol Downstream Peru (an international LPG distributor) and the Ministry of Energy and Mining of Peru, with support from the UNDP (United Nations Development Programme). Stove E was a worn-out appliance with a deteriorated burner obtained from a rural household in Cameroon. These stoves are all commercially available household LPG cookstoves but have different designs, burner types, and air control devices.

(A) Aodian stove. This stove has a single burner, a piezoelectric igniter, and a safety device that cuts off

Table 1. Description of the Stove/Fuel Combinations and Conditions Tested

Series #	Stove	Power level	Burner air adjustment	LPG fuel composition Butane/Propane <sup>a</sup> (% by mass)	Number of valid tests
1	A. Aodian, single burner with two air adjustments	High	Maximum – inner flame Maximum – outer flame ring	20/80	3
2				40/60	5
3				60/40	3
4				80/20	3
5			Minimum – both	40/60	5
6			Maximum – inner flame Minimum – outer flame ring	40/60	3
			Minimum – inner flame Maximum – outer flame ring	40/60	5
7	Low	Maximum – both	40/60	5	
8		B. AOSD, Single burner with one air adjustment	High	Maximum	20/80
9	40/60				5
10	60/40				3
11	80/20				3
12	Medium (air ~ half-open)			40/60	4
13	Minimum		40/60	3	
14	Low		Maximum	40/60	3
15	C. Mikachi, single burner, one air adjustment	High	Maximum	40/60	7
16			Minimum	40/60	3
17		Low	Maximum	40/60	3
18	D. Solgas, double burners no air adjustment	High	None	40/60	3
19		Low	None	40/60	7
20	E. Simcook, three burners, no air adjustment	High	None	80/20	5
21		Low	None	80/20	5
22					

<sup>a</sup>All fuels met US industry standards for composition of minor gases.

the gas supply if the flame fails. The burner has two separate air adjustments for inner and an outer flame rings (Figure S1-a). The stove body is made from stainless steel, and the burner is copper alloy. Brief instructions for adjusting the air control are provided with the stove.

- (B) AOSD stove. This stove has a single burner and a piezoelectric igniter. No safety device is provided to cut off the gas supply if the flame fails. The burner has an inner flame and an outer flame ring, similar to that of Stove A, but the burner has only one air adjustment (Figure S1-b). Materials for Stove B are the same as for Stove A, and brief instructions for adjusting the air control are provided.
- (C) Mikachi stove. This stove has a single burner with one air adjustment and a piezoelectric igniter. No safety device is provided to cut off the gas supply if the flame fails. The structure of the stove body is similar to that of Stove B, but the burner is different (Figure S1-c). The stove body is made from stainless steel, and the burner is cast iron.
- (D) Solgas stove. This stove has two identical burners with no air adjustments, no piezoelectric igniter, and no safety device to cut off the gas supply if the flame fails. Each burner has a separate control to turn the gas on/off and to adjust the cooking power. Since the stove does not include an ignition device, burners must be lit with a match or other source of flame. The stove body is constructed of steel coated with baked enamel. Burner tops are brass, and burner bottoms are cast alloy. The stove was tested by operating only one of the two identical burners (Figure S1-d).

- (E) Simcook stove. This stove was designed with three burners—two identical burners on left and right sides and a smaller burner in the middle (Figure S1-e). This stove has no air adjustments, no piezoelectric igniter, and no safety device to cut off the gas supply if the flame fails. Each burner has a separate control to turn the gas on/off and to adjust the cooking power. The stove body is constructed of steel coated with baked enamel, similar to that of stove D. The stove body and burners were corroded severely after use in a rural household in Cameroon from 2009 to 2016. Due to safety concerns, the stove was replaced by a new one in the rural household and was shipped to our laboratory for emissions testing. This stove provides one example of a badly worn-out LPG appliance, but it may not be representative of all worn-out stoves. The stove was tested by operating the only functional large burner.

**LPG Fuels Tested.** Four different gas compositions with butane/propane weight percentages of 20/80, 40/60, 60/40, and 80/20 were tested using Stoves A and B. The fuels met US industry specifications for olefin and other contaminant gases (Table S1),<sup>29</sup> but results could be different for fuels with more contaminants or different compositions.<sup>30</sup> The presence of the minor constituents may result in different emissions that should be evaluated in the future.

**Testing Protocol.** Two liters of water were heated from ambient to boiling temperature in a flat-bottomed stainless steel pot, and the water continued boiling for the 30 min test duration. Larger pots with lids could be used to boil a larger volume of water, but in the present study, we used a pot size that enabled water to be boiled with no lid. The same pot was

used for all tests. Measurements at the beginning and end of each test included the mass of water in the pot and the mass of fuel in the cylinder. Continuous measurements recorded every 5 s by the data acquisition system included the water temperature, pollutant concentrations, and other test system parameters. Each stove was tested at two power levels by adjusting the rotary valve with a knob on the front of the stove at either minimum (low-power) or maximum (high-power). At least three test replicates were performed for each of the 22 conditions listed in Table 1. Stoves A–C had shutter devices that could be adjusted by the stove user to change the air flow to the burners, and these stoves were tested with various adjustments, as shown in Table 1. Results of a total of 89 valid tests are included in the study presented here.

**Data Analysis.** The total-capture dilution-tunnel method was used to quantify emissions based on continuously measured air flow and pollutant concentrations.<sup>25</sup> A carbon balance check was performed to compare the mass of carbon measured in emissions with the mass of carbon in the fuel, and the acceptance criterion for valid tests was the percent difference based on fuel carbon  $\leq 20\%$ . MCE (modified combustion efficiency), defined as the molar ratio of  $\text{CO}_2/(\text{CO}_2 + \text{CO})$ , was calculated as a proxy for combustion efficiency. TE (thermal efficiency) was calculated as the ratio of useful energy (energy absorbed in the heating and evaporation of the water during the test) divided by fuel energy.

For real-time measurements (all gases and BC), laboratory background concentrations were measured before and after testing each day. For filter-based measurements ( $\text{PM}_{2.5}$ , OC, and EC), laboratory ambient air was sampled and analyzed following the same procedure as emission samples to determine background levels. Average background concentrations were subtracted from total concentrations measured to determine air pollutant emission factors. Detection limits were defined as three times the standard deviation of background concentrations (Table S2).<sup>31</sup> If concentrations were above detection limits, emission factors were calculated on the basis of fuel mass (kg), fuel energy (MJ), and useful energy delivered ( $\text{MJ}_d$ ). If concentrations were below detection, then emission factors were reported to be less than values calculated from detection limits, and those values varied with fuel mass consumption and stove thermal efficiency for the different tests. Differences in the performance metrics under different burning conditions were statistically evaluated using the *t* test or ANOVA (one-way analysis of variance), and the correlation was measured by Pearson's correlation coefficient (*r*) using SPSS (IBM Corp., NY). The significance level of 0.05 was adopted.

## RESULTS

Compared with solid fuel combustion, LPG combustion is generally expected to have lower emissions of most air pollutants. For the 89 tests included in this study, most gases were observed above limits of detection under the various test conditions. The exceptions were  $\text{CH}_4$  for stoves A and D when operated at the low-power level and for stove A when the inner burner's air adjustment was set to minimum (Table S3). Approximately 90% of the  $\text{PM}_{2.5}$ , OC, EC, and BC data were below the corresponding detection limits; thus, emissions of those pollutants were not included in statistical comparisons of results from different fuel compositions, power levels, and stove types. Figure S2 shows emission factor data and corresponding detection limits for  $\text{PM}_{2.5}$ , OC, EC, and BC.

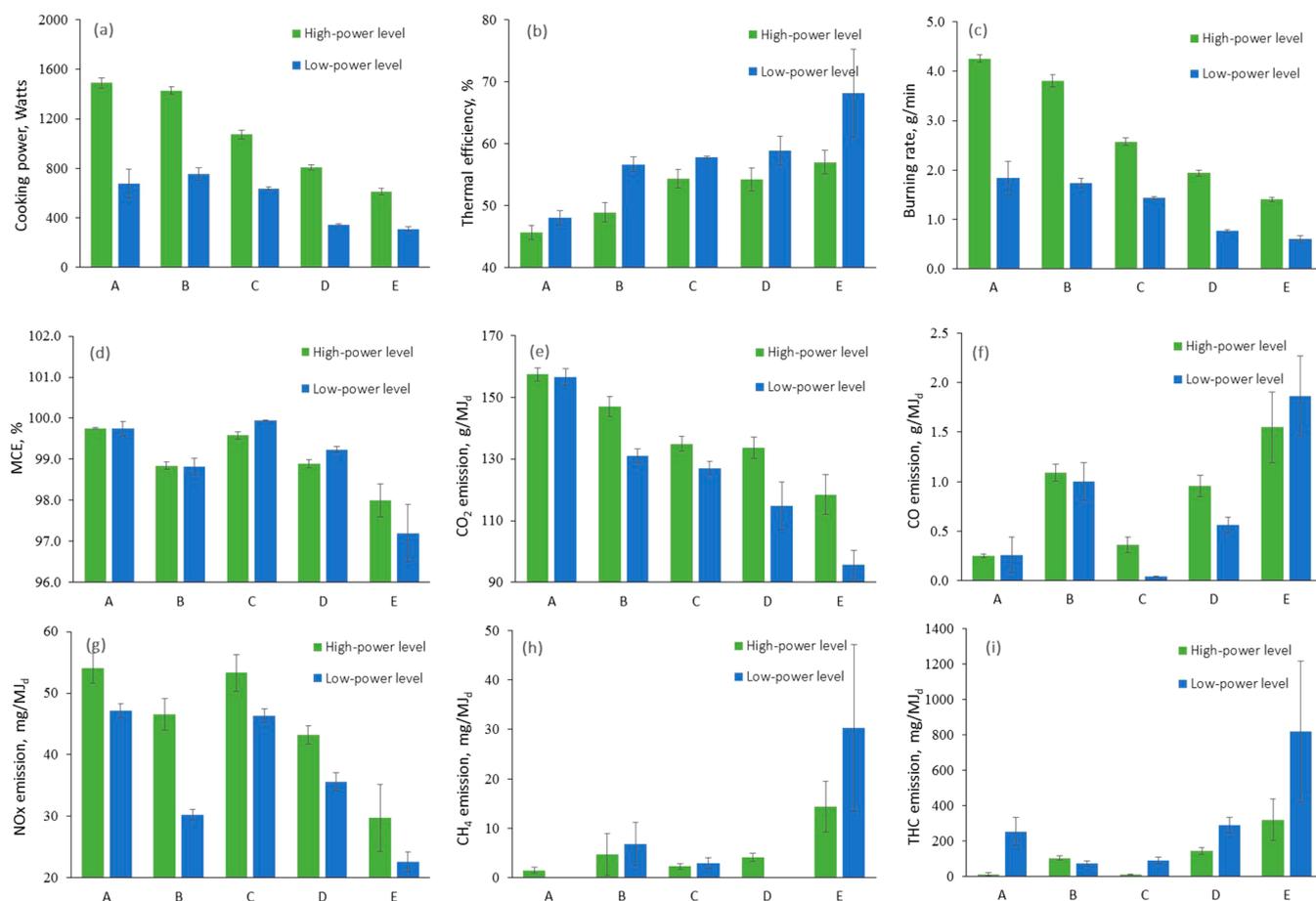
**Influence of Different LPG Fuel Compositions.** Results are provided in Table S4 for the burning of compositionally different LPG fuels in two different stoves. The ANOVA results indicated no significant differences among the four different gas fuel compositions for stove A when considering thermal efficiency, burning rate, cooking power, and most air pollutant emissions.  $\text{CO}_2$  and CO emissions from 40/60, 60/40, and 80/20 fuel blends were significantly higher than emissions from the 20/80 blend ( $p < 0.05$ ), but only by 1.04 and 1.33 times, respectively. Stove B, similarly, shows no significant differences among fuel blends when considering thermal efficiency, burning rate, and cooking power. Compared with the 20/80 and 40/60 blends, the 60/40 and 80/20 blends had comparable but significantly higher  $\text{CO}_2$ , CO, and  $\text{CH}_4$  pollutant emissions—1.03, 1.25, and 1.96 times higher, respectively ( $p < 0.05$ ).

**Difference between the High- and Low-Power Levels.** The difference between the high- and low-power levels was studied using the same 40/60 blend fuel.  $\text{CO}_2$  concentrations measured in the dilution tunnel are graphed with water temperature profiles during high- and low-power level tests in Figure S3. Detailed results for high- and low-power are provided in Table S5. Results are consistent for the five stoves—stoves operating at low-power level have less cooking power, a slower burning rate, and significantly higher thermal efficiency compared with high-power operation. MCE values compared at both the high- and low-power levels show no significant difference for Stoves A, B, and E, but for stoves C and D, MCEs were significantly different at low-power. Because  $\text{TE} \approx \text{MCE} \times \text{HTE}$  (heat transfer efficiency),<sup>25</sup> the higher thermal efficiency for the low-power is due mainly to enhanced heat transfer efficiency with a slower burning rate.

Regarding air pollutant emission factors based on useful energy delivered, the difference between high- and low-power levels varied for different stoves and pollutants.  $\text{CO}_2$  emission factors were slightly less at low-power for all stoves, with significant differences for Stoves B, C, D, and E. CO emission factors were similar at high- and low-power levels for Stoves A, B, and E but were significantly less at the low-power level for Stoves C and D (less by 89% and 40%, respectively). THC emissions during low-power operation were significantly greater than those observed during high-power operation for Stoves A, C, D, and E. THC emissions were significantly decreased, however, for Stove B.  $\text{CH}_4$  emission factors were apparently higher at low-power operation for Stoves B, C, and E, but only stove C had a difference that was statistically significant. No comparisons were possible for Stoves A and D because  $\text{CH}_4$  was less than the detection limit at the low-power level.  $\text{NO}_x$  emissions were significantly less at low-power for all stoves (decreased by 13–35%), likely due to lower combustion temperatures.

**Influence of Burner Air Adjustments.** The air supply to the burner, more specifically the air/fuel ratio, is critical for combustion and consequent emissions of air pollutants. A comparison of emissions results for different burner air adjustments is provided in Table S6. Thermal efficiency, burning rate, and cooking power did not show significant differences with the burner air adjustments for the three tested stoves (stoves D and E did not have air adjustment devices).

Yellow flames indicate the presence of incandescent soot particles, while blue flames indicate more complete combustion.<sup>32</sup> In this study, blue flames were observed visually during testing of all stoves under most test conditions, and yellow flames were only observed for stove A when the air for the



**Figure 3.** Comparison of cooking power (a), thermal efficiency (b), burning rate (c), modified combustion efficiency (MCE, d), and emission of CO<sub>2</sub> (e), CO (f), NO<sub>x</sub> (g), CH<sub>4</sub> (h), and THC (total hydrocarbon (i)) for the five stoves tested (stoves A–E). Results shown are means, and standard deviations are indicated by the error bars.

outer flame ring was at the minimum setting (series 5 and 6 in Table 1). Stove A was the only stove with two separate air adjustments for the inner and outer flame rings. In the yellow flame cases (series 5 and 6), MCE was lower and CO emissions were approximately 1.6 times higher than in tests with blue flames observed. NO<sub>x</sub> emissions show no significant differences with burner air settings. THC emissions were also apparently higher with yellow flames, but the difference is insignificant due to the high variability in these emissions. In tests where yellow flames were observed, EC and BC were detected in all test replicates—with average emission factors of  $0.40 \pm 0.23$  and  $0.64 \pm 0.33$  mg/MJ<sub>d</sub>, respectively; but in tests when blue flames were observed, EC was not detectable and BC was only detected in two tests (Figure S2). PM<sub>2.5</sub> and OC were detected in 38% of the samples from the yellow flame cases, with emission factors of 1.6–2.6 and 0.44–0.98 mg/MJ<sub>d</sub>, respectively.

For Stoves B and C, when the burner air controller was adjusted from the maximum to the minimum setting, the MCE decreased slightly and this decrease is significant (we note that the flames were still blue at minimum setting for these stoves). CO, THC, and CH<sub>4</sub> emissions increased significantly. NO<sub>x</sub> emissions decreased significantly only for stove C, from  $53.3 \pm 3.0$  to  $48.7 \pm 0.5$  mg/MJ<sub>d</sub>.

**Comparison of Five Different Stoves.** The results in Figure 3 suggest that stove design and condition are important in terms of stove efficiency and air pollutant emissions. The

thermal efficiency, burning rate, cooking power, MCE, and gas emissions varied significantly among the five stoves tested. Cooking power (Figure 3a) and fuel burning rate (Figure 3c) are in descending order with Stoves A > B > C > D > E; however, the thermal efficiency (Figure 3b) shows the opposite order. Cooking power correlated positively with fuel burning rate ( $r = 0.99$ ,  $p < 0.05$ ), as expected, and these two parameters correlated negatively with thermal efficiency ( $r = -0.82$  and  $-0.86$ , respectively,  $p < 0.05$ ). The MCE values (Figure 3d) for Stoves A and C were higher than the values for Stoves B and D, and the lowest MCE values were for Stove E. Stoves A and C had lower CO emissions (Figure 3f) but relatively higher NO<sub>x</sub> emissions (Figure 3g) than Stoves B, D, and E. CH<sub>4</sub> (Figure 3h) and THC (Figure 3i) emissions were found to be much higher for the deteriorated Stove E compared with the other new stoves. Maximum cooking power varied by a factor of nearly two among the stoves tested. Thermal efficiency varied with a difference of approximately 10% between lowest and highest performing stoves.

The difference in burner type across the five tested stoves may be an important factor affecting the observed differences in heat transfer and burning efficiency. There are many different types of burners with distinct designs often made from different materials. Thermal efficiency was reported to increase from 48% to 52% when a cast iron burner was replaced with a brass burner.<sup>33</sup> The burner of Stove D in the present study was made from brass, and it had a slightly higher thermal efficiency than

**Table 2. Summary of Air Pollutant Emission Factors on the Basis of Fuel Mass (kg), Fuel Energy (MJ), and Useful Energy Delivered (MJ<sub>d</sub>) and Emission Rates on the Basis of Time (min) for Household LPG Cookstoves<sup>a</sup>**

	units	range	mean ± SD	median		units	range	mean ± SD	median
CO <sub>2</sub> (n = 89)	g/kg	2656–3584	3302 ± 144	3319	PM <sub>2.5</sub> (n = 9)	mg/kg	26–141	53 ± 37	35
	g/MJ	58–78	72 ± 3	72		mg/MJ	0.57–3.1	1.2 ± 0.8	0.76
	g/MJ <sub>d</sub>	92–163	142 ± 17	147		mg/MJ <sub>d</sub>	1.2–6.5	2.4 ± 1.6	1.6
	g/min	1.6–15	9.8 ± 4.1	12		mg/min	0.11–0.61	0.20 ± 0.16	0.14
CO (n = 89)	g/kg	0.97–75	19 ± 15	17	OC (n = 9)	mg/kg	9.6–108	29 ± 32	16
	g/MJ	0.021–163	0.42 ± 0.33	0.38		mg/MJ	0.21–2.4	0.62 ± 0.68	0.36
	g/MJ <sub>d</sub>	0.036–2.3	0.77 ± 0.55	0.60		mg/MJ <sub>d</sub>	0.44–5.0	1.2 ± 1.4	0.66
	g/min	0.001–0.15	0.049 ± 0.038	0.034		mg/min	0.039–0.47	0.10 ± 0.14	0.055
CH <sub>4</sub> (n = 72)	g/kg	0.016–1.5	0.15 ± 0.25	0.064	EC (n = 8)	mg/kg	1.4–16	8.5 ± 4.9	7.7
	mg/MJ	0.34–33	3.3 ± 5.4	1.4		mg/MJ	0.031–0.35	0.18 ± 0.11	0.17
	mg/MJ <sub>d</sub>	0.72–47	5.6 ± 8.2	2.6		mg/MJ <sub>d</sub>	0.070–0.78	0.40 ± 0.23	0.36
	mg/min	0.067–1.2	0.27 ± 0.25	0.18		mg/min	0.006–0.066	0.035 ± 0.020	0.031
THC (n = 89)	g/kg	0.086–36	3.6 ± 6.1	2.2	BC (n = 9)	mg/kg	2.1–25	11 ± 8	7.8
	mg/MJ	1.9–776	78 ± 132	48		mg/MJ	0.045–0.54	0.24 ± 0.17	0.17
	mg/MJ <sub>d</sub>	4.1–1144	130 ± 196	94		mg/MJ <sub>d</sub>	0.10–1.2	0.53 ± 0.37	0.37
	mg/min	0.36–21	5.8 ± 5.1	5.4		mg/min	0.009–0.10	0.045 ± 0.031	0.031
NO <sub>x</sub> (n = 89)	g/kg	0.58–1.4	1.1 ± 0.2	1.1					
	mg/MJ	13–31	23 ± 4	24					
	mg/MJ <sub>d</sub>	21–60	46 ± 9	48					
	mg/min	0.37–5.3	3.3 ± 1.5	3.8					

<sup>a</sup>Results are range, mean ± standard deviation (SD), and median. Data below the limits of detection are excluded in these summarized results. The limits of detection are listed in Table S2. Sample size (n) is provided. THC: total hydrocarbon; PM<sub>2.5</sub>: particles with aerodynamic diameter ≤2.5 μm; OC: organic carbon in PM<sub>2.5</sub>; EC: elemental carbon in PM<sub>2.5</sub>; BC: black carbon.

Stove C which had a cast iron burner; however, since the stove design was different for these two stoves, the higher thermal efficiency may not be solely explained by the burner material difference. Previous studies also showed that a swirling flow burner had higher thermal efficiency but increased CO emissions compared to a conventional radial flow burner.<sup>34,35</sup> The overall efficiency may be improved notably by using catalytic combustion in a ceramic matrix;<sup>36</sup> however, the technology is not typical at present for household cooking devices.

As discussed above, the performance of LPG stoves can vary with stove types and burning conditions; however, for all tests, the overall mean and standard deviation for thermal efficiency was 51 ± 6%, and emissions of CO<sub>2</sub>, CO, THC, CH<sub>4</sub>, and NO<sub>x</sub> were 142 ± 17, 0.77 ± 0.55, 130 ± 196, 5.6 ± 8.2, and 46 ± 9 mg/MJ<sub>d</sub>, respectively. Pollutant emission factors on the basis of fuel mass (kg), energy (MJ), and useful energy (MJ<sub>d</sub>) as well as emission rates on the basis of time (min) are summarized in Table 2. The COVs (coefficients of variation) for CO<sub>2</sub>, CO, THC, CH<sub>4</sub>, and NO<sub>x</sub> emission factors were 12%, 72%, 151%, 147%, and 20%, respectively. In the repeated tests under the same conditions, the COVs were in the range of 0.2–6.7%, 1.4–70%, 2.0–80%, 6.8–62%, and 1–18% for CO<sub>2</sub>, CO, THC, CH<sub>4</sub>, and NO<sub>x</sub>, respectively, and were generally lower than the overall COVs when combining results from all tests. This indicates that pollutant emission factors varied much more between stove types and other study parameters than between testing replicates under the same conditions.

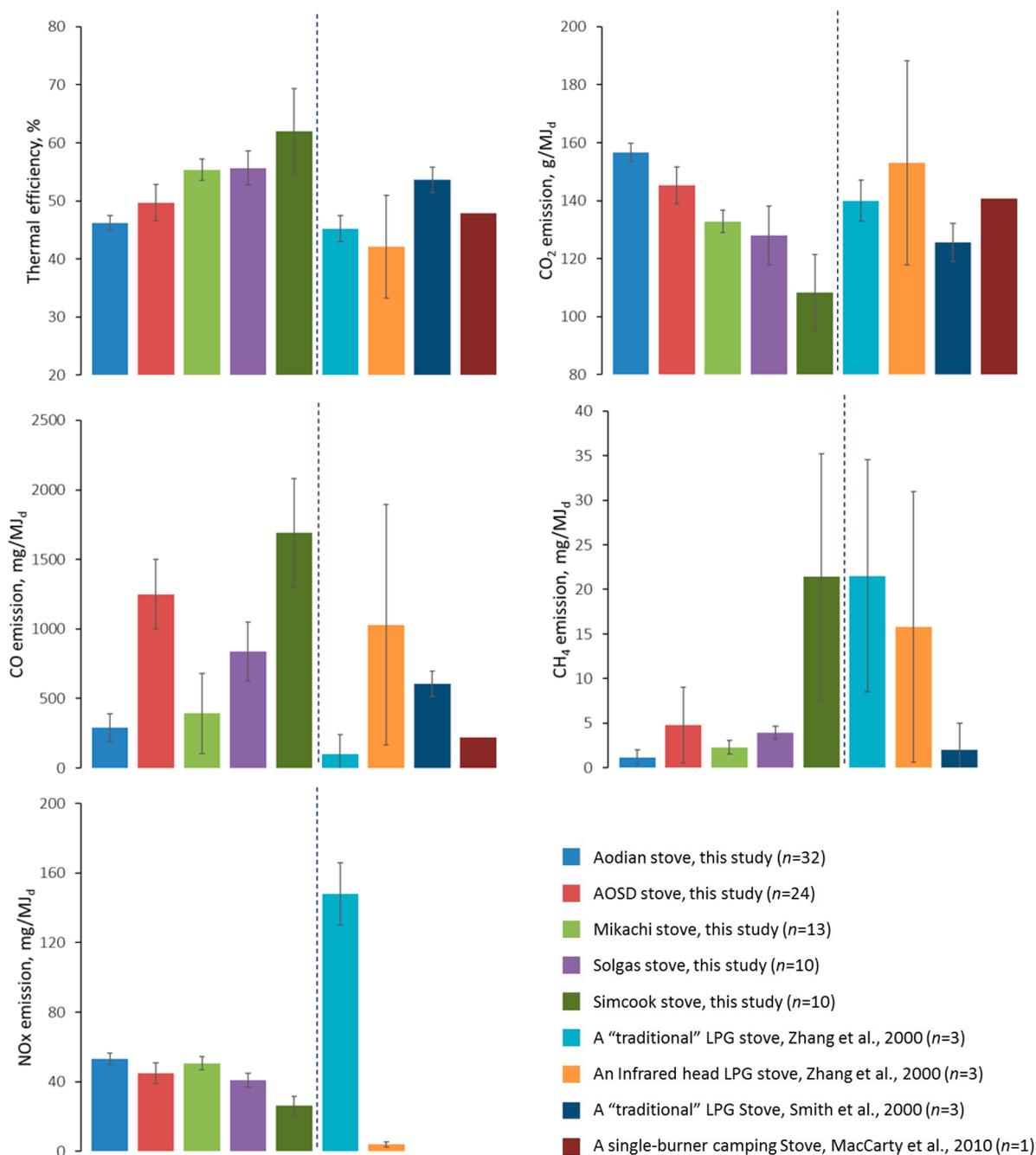
Approximately 90% of the PM<sub>2.5</sub>, OC, EC, and BC data were below their corresponding detection limits. For those data above the detection limits, PM<sub>2.5</sub>, OC, EC, and BC emission factor ranges were 1.2–6.5, 0.44–5.0, 0.070–0.78, and 0.10–1.2 mg/MJ<sub>d</sub>, with overall means and standard deviations of 2.4 ± 1.6, 1.2 ± 1.4, 0.40 ± 0.23, and 0.53 ± 0.37 mg/MJ<sub>d</sub>, respectively. Most of these were found in the yellow flame cases

when the air supply was reduced. A previous emission study on gas furnaces and heaters reported that particle emissions were similar between yellow and blue fires (0.28 ± 0.11 and 0.28 ± 0.18 mg/MJ, respectively, on the basis of fuel energy); however, emissions would increase by a factor of 30 (9.2 ± 3.5 mg/MJ) in a luminous white flame.<sup>37</sup> In the present study, a luminous white flame was not observed under any test conditions.

## DISCUSSION

**Comparison with Previous Studies.** Results are compared to available literature studies for LPG cookstoves.<sup>14–17</sup> Note that different protocols and methodologies were used in these studies which may also contribute to the difference in results, in addition to the different stoves that were tested. As shown in Figure 4, the thermal efficiencies (46–62% across all stoves included in this study) were similar to, or somewhat higher than, the range of reported efficiencies in the literature (42–54%). Cooking vessels used in both literature and the present tests were typical flat-bottomed pots made from stainless steel. Thermal efficiency can vary with different pots.<sup>33</sup> Pots with heat-transfer fins on the bottom are widely available in some countries and commonly used in restaurant kitchens, but these pots may be too expensive at present for many households in low- and middle-income areas.

CO<sub>2</sub> emissions in the present study were also comparable to those in the three cited studies (108–157 g/MJ<sub>d</sub> versus 126–153 g/MJ<sub>d</sub>, respectively). CO emissions in both the present study and in the literature showed large differences between different stoves, and generally the CO emission ranged from 100 to 1700 mg/MJ<sub>d</sub>. NO<sub>x</sub> emissions from two stoves in the literature were 148 ± 18 and 4.1 ± 1.4 mg/MJ<sub>d</sub> for a conventional stove and for a stove with an infrared head, respectively.<sup>14</sup> Our results, ranging from 27 ± 5 to 53 ± 3 mg/MJ<sub>d</sub>, were in the middle of the range of these data sets. CH<sub>4</sub> emissions for the four well-functioning stoves in our study were



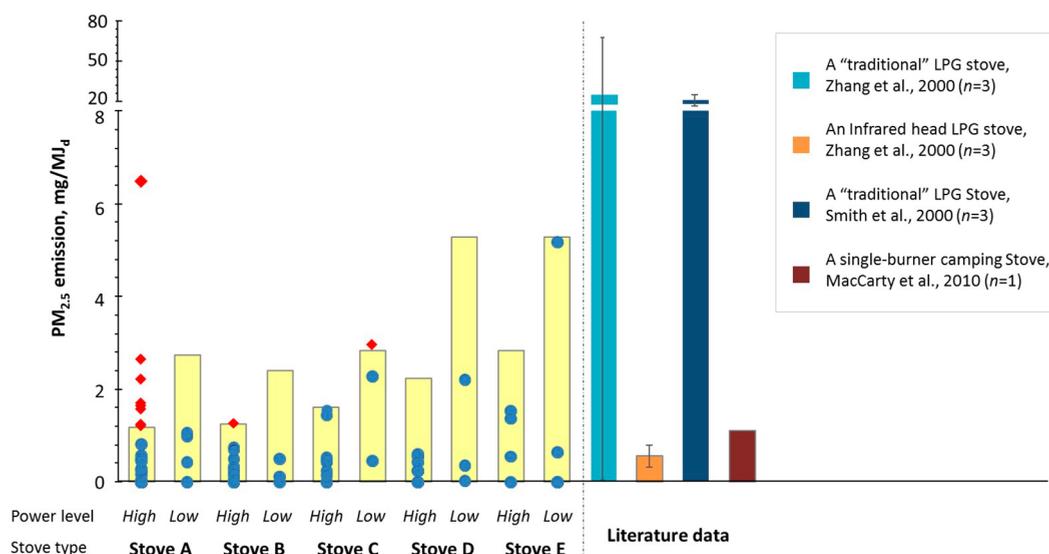
**Figure 4.** Comparison of thermal efficiency and air pollutant emission factors from LPG cookstoves with other studies in the literature (refs 14, 15, and 17). Data shown are means and standard deviations.

in 1.5–5.0 mg/MJ<sub>d</sub>, similar to the reported result of 2.0 mg/MJ<sub>d</sub> for a traditional two-burner LPG stove in India.<sup>15</sup> For degraded Stove E, CH<sub>4</sub> levels were as high as 21 ± 14 mg/MJ<sub>d</sub>. Tests by Zhang et al. (2000)<sup>14</sup> on two typical LPG stoves in China also reported high CH<sub>4</sub> emissions of 23 and 16 g/MJ<sub>d</sub>, respectively.

For particle emissions (Figure 5), some previous studies reported TSP (total suspended particles) instead of PM<sub>2.5</sub>,<sup>14,15</sup> but the comparison here is acceptable as particles from LPG combustion are typically less than 2.5 μm.<sup>18</sup> Past studies have reported a wide range of particle emissions. A value of 0.54 ± 0.24 mg/MJ<sub>d</sub> was reported for an LPG-fueled infrared head stove,<sup>14</sup> and 1.1 mg/MJ<sub>d</sub> was reported for a propane-fueled single-burner camping stove tested using a light-scattering

sensor.<sup>17</sup> Two reported values of 24.9 ± 42.8 by Zhang et al.<sup>14</sup> and 20.9 ± 3.8 mg/MJ<sub>d</sub> by Smith et al.<sup>15</sup> were much higher. The present study fell near the low end of this observed range with only 10% of the tests exceeding the PM<sub>2.5</sub> detection limit. For Stoves A, B, and C, most PM<sub>2.5</sub> emission factors were below 1.5 mg/MJ<sub>d</sub> at the high-power level and below 3.0 mg/MJ<sub>d</sub> at the low-power level. For Stoves D and E, most PM<sub>2.5</sub> emission factors were below 3.0 mg/MJ<sub>d</sub> at the high-power level, and below 5.5 mg/MJ<sub>d</sub> at the low-power level. For those data above the detection limit, the average PM<sub>2.5</sub> emission factor was 2.4 ± 1.6 mg/MJ<sub>d</sub>.

Besides the three studies referred to in Figure 5, Habib et al.<sup>16</sup> also measured PM<sub>2.5</sub> and its chemical properties from LPG burning in India. Results were reported on the basis of fuel



**Figure 5.**  $\text{PM}_{2.5}$  emission factor data from the present study and literature studies (refs 14, 15, and 17). Emission factors calculated from limits of detection for each stove under high and low power levels are shown as light yellow bars. Data points below the corresponding detection limits are shown as blue dots, while those above the limit are shown as red diamonds.

mass. Since fuel LHV (lower heating value) and TE were not reported in that study, the results cannot be converted to emissions per useful energy and compared in Figure 5. Figure S4 provides a comparison of PM emission factors on the basis of fuel mass. The  $\text{PM}_{2.5}$  emission factor from Habib et al. was 200 mg/kg, and the EC and OC emission factors were 8 and 52 mg/kg, respectively.<sup>16</sup> The EC and OC emission factors were in the range of the present study (1.4–16 and 9.6–108 mg/kg for EC and OC, respectively). The  $\text{PM}_{2.5}$  emission factor was higher than our results (26–141 mg/kg), but fell into the reported range in the literature (Figure S4), and was lower than the past results of  $524 \pm 901$  mg/kg by Zhang et al.<sup>14</sup> and  $514 \pm 93$  mg/kg by Smith et al.<sup>15</sup>

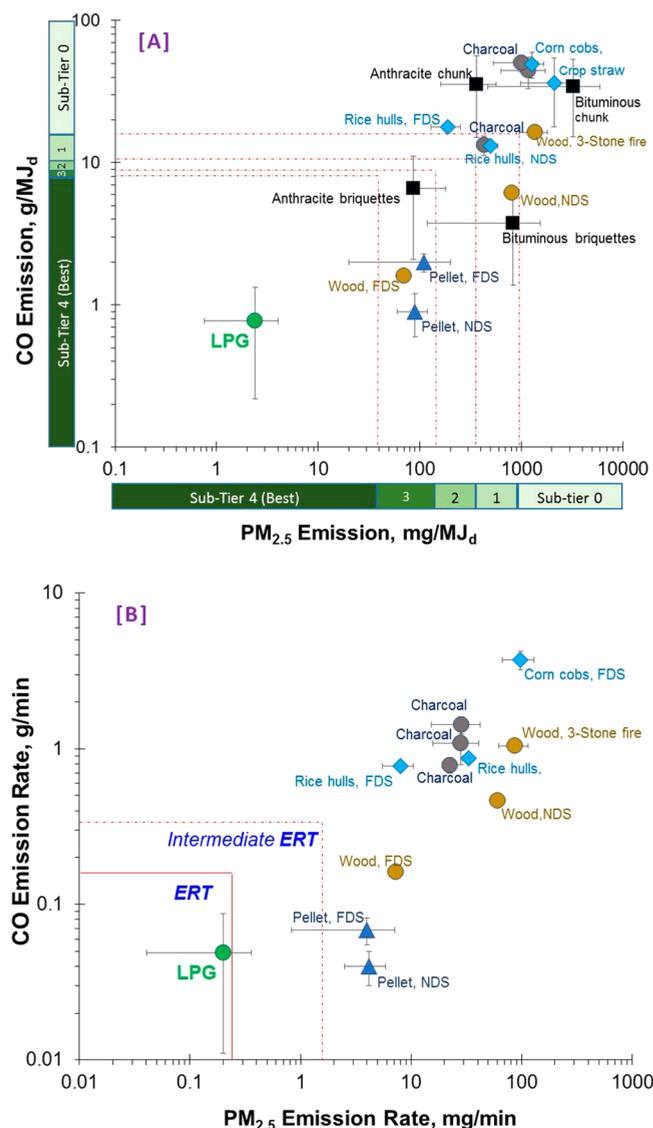
**Evaluation of LPG Cookstove Performance.** According to the ISO IWA guidelines for CO and  $\text{PM}_{2.5}$  emissions,<sup>24</sup> stoves are rated Sub-Tier 4 (best rating) when  $\text{PM}_{2.5} \leq 41$  mg/ $\text{MJ}_d$  and  $\text{CO} \leq 8$  g/ $\text{MJ}_d$ . Results for LPG cookstoves tested under all conditions were clearly within Tier 4 for both CO and  $\text{PM}_{2.5}$  emissions, even for Stove E which was badly worn-out from daily use over approximately seven years in a rural household. The maximum  $\text{PM}_{2.5}$  emission factor in the present study was 6.7 mg/ $\text{MJ}_d$ , well below the Tier 4 limit of 41 mg/ $\text{MJ}_d$ . Previous studies of biomass cookstoves showed that CO and  $\text{PM}_{2.5}$  emissions during the cold-start, high-power test phase ranged from 1.0 to 40 g/ $\text{MJ}_d$  and 60–1400 mg/ $\text{MJ}_d$ , respectively.<sup>25</sup> For coal cookstoves, CO and  $\text{PM}_{2.5}$  have been reported in the range of  $\sim 4$ –40 g/ $\text{MJ}_d$  and  $\sim 100$ –3000 mg/ $\text{MJ}_d$ .<sup>38</sup> Thus, as expected, the deployment of LPG-fueled cookstoves would result in large reductions in CO and  $\text{PM}_{2.5}$  emissions compared to most typical solid fueled stoves (Figure 6a). Note that in Figure 6,  $\text{PM}_{2.5}$  emissions for LPG include only results above the detection limits.

Emission rates can provide important information about potential health risks, and the ERTs (emission rate targets) and Intermediate ERTs for CO and  $\text{PM}_{2.5}$  from vented and unvented stoves are recommended in the WHO (World Health Organization) guidelines on household air pollution.<sup>39</sup> CO emission rates in the present study ranged from 0.0014 to 0.15 g/min, which were within the WHO ERT of 0.16 g/min for

unvented stoves. For  $\text{PM}_{2.5}$ , 90% of the tests had emission rates  $< 0.11$  mg/min, while the remaining tests with data above the detection limit had an emission rate range of 0.11–0.61 mg/min, with a mean and standard deviation of  $0.20 \pm 0.16$  mg/min.  $\text{PM}_{2.5}$  emission rates were clearly within the WHO intermediate ERT of 1.75 mg/min, and most data points were within the final ERT of 0.23 mg/min, as well (Figure 6b).

**Implications, Limitations, and Future Work.** In this study, 89 laboratory tests were performed to evaluate efficiency and air pollutant emissions from five household LPG cookstoves. The influence of fuel composition, stove power level, burner air adjustment, and stove condition was investigated. Larger differences in performance were observed among the different stove designs tested compared with the relatively smaller variations due to the operational variables mentioned above. The study data will be useful in developing emission inventories and evaluating impacts of LPG interventions on air quality and human health. The study confirmed high efficiency and low emissions of LPG cookstoves. Relative to typical solid fuel stoves, a significant reduction in air pollutant emissions and an obvious improvement in indoor air quality can be expected from the adoption of LPG cookstoves. These results may increase confidence in the ongoing LPG intervention programs in many developing countries.

Limitations of the present study and the need for future laboratory and field investigations are acknowledged. Commercially available LPG cookstoves were tested without any modifications, and some stove design aspects were not evaluated in the present study, such as primary aeration, burner type (materials and shape), and loading height (the vertical distance from the top of the burner port to the bottom of the vessel).<sup>34,35,40</sup> Stoves were tested at the gas pressure of 2.8 kPa, as specified by the manufacturers. Pressure inside the cylinder varies with temperature, but delivered gas pressure remains nearly constant with a properly functional pressure regulator. It was reported that when the gas supply pressure was increased, thermal efficiency decreased and CO concentrations increased due to increased flame impingement on the pot surface.<sup>30,40</sup> As mentioned above, the LPG fuel in this study met U.S. industry



**Figure 6.** CO and PM<sub>2.5</sub> emission factors per useful energy delivered (A) and emission rates (B). Emission factors are compared to ISO IWA performance guidelines, and emission rates are compared to the WHO emission rate targets (ERTs) for unvented stoves. PM<sub>2.5</sub> emissions reported for LPG are only from results above the detection limits in the present study. For comparison, selected fuel-stove combinations from previously published data are also included in the figure. The literature data are cited to indicate general ranges of CO and PM<sub>2.5</sub> emissions from different solid fuel cookstoves. Data for wood, pellet, rice hulls, and charcoal in natural- or forced-draft stoves (NDS and FDS) were from Jetter et al.,<sup>25</sup> and emissions from coal cookstoves were from Shen.<sup>38</sup>

specifications, while in other LPG supplies, the presence of minor constituents may result in different emissions that should be evaluated. Future laboratory and field studies are needed to characterize performance, including cases such as those associated with malfunctioning stoves, malfunctioning gas pressure and flow regulators, and contamination of fuels and stove burners in rural homes.

There are many different LPG stoves available in world markets, and the performance could be different compared with the five stoves tested in this study. Given the low detection frequency of PM<sub>2.5</sub> in the present study, the overall average PM<sub>2.5</sub> emissions from LPG would likely be much lower.

However, some high-emission events occurred with emission rates that could be slightly higher than the WHO final ERT. For example, one test in the present study had a relatively high PM<sub>2.5</sub> emission rate of 0.61 mg/min, the reason for which was unclear. Future work on different LPG stoves is encouraged, and efforts to increase limits of detection (e.g., by using a high-volume sampler to capture more mass of PM<sub>2.5</sub>; having lower levels and less variations of background concentrations) would be useful.

Note that this study does not consider upstream emissions from the LPG fuel cycle, for example, from refineries, gas wells, or renewable sources. Potential problems like LPG leaks in the distribution and storage systems that can lead to substantial regional air quality impacts should be considered in large-scale LPG intervention programs as well.

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b05155.

Pictures of stoves; CO<sub>2</sub> and water temperature profiles during the high- and low-power levels; comparison of PM emission factors on the basis of fuel mass (mg/kg) from the present study and literature results; concentrations of minor constituents in the LPG fuel; calculated values from limits of detection for each test; and data of burning rates, cooking power, thermal efficiency, MCE, and gas emission factors under different testing conditions (PDF)

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### Notes

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