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Pollutant Emissions and Energy Efficiency under Controlled Conditions for Household Biomass Cookstoves and Implications for Metrics Useful in Setting International Test Standards

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ABSTRACT: Realistic metrics and methods for testing household biomass cookstoves are required to develop standards needed by international policy makers, donors, and investors. Application of consistent test practices allows emissions and energy efficiency performance to be benchmarked and enables meaningful comparisons among traditional and advanced stove types. In this study, twenty-two cookstoves burning six fuel types (wood, charcoal, pellets, corn cobs, rice hulls, and plant oil) at two fuel moisture levels were examined under laboratory-controlled operating conditions as outlined in the Water Boiling Test (WBT) protocol, Version 4. Pollutant emissions (carbon dioxide, carbon monoxide, methane, total hydrocarbons, and ultrafine particles) were continuously monitored. Fine particle mass was measured gravimetrically for each WBT phase. Additional measurements included cookstove power, energy efficiency, and fuel use. Emission factors are given on the basis of fuel energy, cooking energy, fuel mass, time, and cooking task or activity. The lowest $PM_{2.5}$ emissions were 74 mg $MJ_{delivered}^{-1}$ from a technologically advanced cookstove compared with 700-1400 mg $MJ_{delivered}^{-1}$ from the base-case open 3-stone cookfire. The highest thermal efficiency was 53% compared with 14-15% for the 3-stone cookfire. Based on these laboratory-controlled test results and observations, recommendations for developing potentially useful metrics for setting international standards are suggested.

INTRODUCTION

Solid fuel combustion emissions from household cooking and heating are a leading risk factor for disease in the developing world, accounting for approximately 4% of all lost healthy life years and some 2 million premature deaths in low- and middle-income countries[1]. Demand for fuelwood resources for household energy impacts terrestrial ecology and land-use patterns in a number of regions within the developing world, and fuel gathering typically requires many hours per week for poor populations[2]. Potential climate effects are of concern due to the greenhouse gas and carbonaceous aerosol emissions from household combustion of biomass fuel[3]. Evidence suggests that widespread deployment of cookstoves with energy and combustion efficiency improvements over traditional technology could potentially help mitigate adverse human health, energy, and climate consequences[4]. Studies of stoves with chimneys show some success in reducing exposures and health impacts, but also indicate that additional reduction in emissions could potentially achieve even greater benefits[5]. However, wide-scale adoption of cookstoves built with improved combustion technologies and low emissions faces substantial challenges, including the lack of widely available and accepted cookstove emissions and energy efficiency standards and testing protocols[6]. Such standards are required for (i) informing governments, donors, and investors interested in promoting and supporting only high-quality stoves, (ii) improving comparisons among fuels and stoves operating under pertinent task-, energy efficiency-, and combustion-related test variables, and (iii) developing certification procedures, performance benchmarks, and meaningful test infrastructure for the global cookstove market. Standards can provide incentive for stove developers to innovate and improve performance. Standards are not developed in this study but may be developed through a standards organization. Metrics are suggested for possible use in standards, as discussed below.

A variety of protocols and metrics are used to evaluate cookstoves and quantify their combustion emissions. Many studies utilize the "hood method" to capture, dilute, and measure air pollutant emissions from cookstoves[7-20], while some measure the emissions directly from the flue if there is a chimney[17-20] or from a chamber after re-directing the emissions[21-25]. These studies certainly provide a valuable body of knowledge, but the numerous test procedures have led to inconsistencies in reporting, making the sparse published data difficult to compare from one laboratory to the next. Moreover, the combustion and fuel conditions, chemical and physical analysis techniques, reporting metrics, and pollutant types measured in these studies vary substantially. This general lack of consistency has hampered the development of sound policies regarding cookstove use and dissemination. Further complicating the picture is that the emissions from cookstove testing under field and laboratory conditions can differ[26-28].

The development of international cookstove standards will require some degree of laboratory control to rapidly and accurately assess energy- and task-specific emissions performance as cookstove design and technology improve. Many earlier emissions characterization efforts successfully applied controlled laboratory conditions to examine cookstoves from Asia[8-12,17-21,29-33] , South Africa[7], and Guatemala[22], where populations rely heavily on their use. More recent controlled laboratory experiments were conducted to investigate the effects of fuel species, fuel combinations and fuel moisture content on stove emissions and energy efficiency[13-15,23-25]. Multiple stove design technologies were examined in these experiments, and test results for fifty stoves were compiled for the purpose of defining composite performance benchmarks[16], but a different approach is recommended here, as described below.

The present study provides a more extensive analysis of emissions and fuel use from a wider range of newer cookstove technologies than past studies. Twenty-two cookstoves burning six fuels (cookstove dependent) at two moisture content levels are examined under laboratory-controlled operating conditions. Pollutant emissions are sampled using established hood and dilution methods. Real-time measurements of carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), total hydrocarbon (THC), fine particulate matter, and ultrafine particle (UFP) emissions are provided. In the interest of developing a novel and versatile emissions database for cookstoves, emissions rates and factors are calculated on the basis of cooking energy delivered, cooking task, fuel energy and fuel mass. Cooking power, energy efficiency, and fuel use are also calculated. Using these laboratory-controlled test results and observations as a basis, useful metrics for developing and setting international standards for cookstove emissions and energy efficiency are discussed.

MATERIALS AND METHODS

Cookstove Systems. For this study, a cookstove system was defined by the cookstove type (including chimney, if so equipped), fuel (composition, moisture content, and size), cooking pot, pot skirt (device for improving heat transfer) if available from the stove manufacturer, and operating procedure. The twenty-two cookstove types, six fuels, moisture contents (low and high), cooking pot water volumes, and combustion chamber materials (metal or ceramic) are listed in Table 1. A total of forty-four system combinations were tested for the present study (see the Supporting Information for more details). Some low-power stoves did not consistently boil 5L of water—the WBT-specified default volume. For these stoves, a 2L pot was used instead. Two cookstoves were equipped with chimneys (~2m height). Natural-draft (also termed natural convection) stoves dominated the cookstove matrix; four forced-draft (fan-provided air) stoves were also tested. Twelve stoves with batch fuel loading (e.g., charcoal stoves) required less time for tending than others that required manual fuel feeding. Four stoves were variations of the “rocket” stove design[13], and eight were variations of the “gasifier” design[34]. The only traditional cooking system tested was a 3-stone cookfire. This particular system is treated as the base-case in this study because it is traditionally the most widely used. For the “carefully tended” 3-stone cookfire, fuelwood sticks were arranged in a radial pattern with the fire at the center, and sticks were continually fed into the center so that the ends of the sticks consistently burned. In the “minimally tended” mode, fuel wood was loaded in batches approximately every 10 minutes, and the fire was untended between loadings. Apart from the 3-stone cookfire, each cookstove was operated in one mode generally following manufacturers’ instructions. Table 1 provides identifiers that refer to cookstove photos and descriptions in the Supporting Information.

Fuels. Fuels were selected based on the stove type and typical field conditions. Fuel types include wood, charcoal, pellets, corn cobs, rice hulls and plant oil (Table 1). For wood-fired stoves, red oak (*Quercus rubra*) sticks of ~10% and ~30% moisture (wet basis) were used. Wood fuels sometimes required light processing including cutting or chopping. Fuel variables are described in the Supporting Information. Fuel moisture content was measured using ASTM Standard Method D4442-07[35]. Fuel heat of combustion was measured using ASTM Standard Method ASTM D5865-10[36]. Per WBT specification, the lower heating value of each fuel was used (see the Supporting Information).

Test Protocol. The WBT protocol (Version 4)[37] was used to determine cookstove power, energy efficiency, and fuel use. Pollutant emissions were simultaneously measured and reported for each of the three WBT test phases: (1) high-power, cold-start; (2) high-power, hot-start; and (3) low-power, simmer. Phases (1) and (2) were defined by the duration between fire ignition and the water boiling point. Phase (1) began with the cookstove, pot, and water at ambient temperature. Phase (2) immediately followed with the cookstove hot but the pot and water at ambient temperature. Phase (3) was defined by a 30-minute time period with the nominal water temperature maintained at 3°C below the boiling point. A modified procedure was used for charcoal stoves[15] (see the Supporting Information). Two wood-fueled rocket stoves (Envirofit G-3300 and StoveTec GreenFire) were tested at an additional medium-power level. The power level was controlled by simply changing the fuel feed rate. The WBT protocol specifies that the cooking pot be uncovered during testing. Results are reported as averages with standard deviations for the tests performed in triplicate (or more).

Cookstove Emissions Testing Facility. A schematic diagram of the emissions testing system and a thorough description of the facility are provided in the Supporting Information. Briefly, stove emissions were collected into a stainless steel hood connected to a dilution tunnel (~47 m^3 min^{-1}), from which pollutant emissions were sampled. An induced-draft blower maintained negative pressure in the entire system and provided filtered dilution air and hood air flows. A second stage of dilution (~1:10) was provided by a modified dilution sampling system[38,39], which was required for the Scanning Mobility Particle Sizer (SMPS) and certain optical and carbonaceous aerosol measurements to be reported in a subsequent publication. In this study, total emissions—those from the stove body and flue for stoves with chimneys—were measured for all cookstove systems.

Emissions Characterization. CO, CO_2, THCs, and CH_4 were continuously monitored with infrared and flame ionization detector (FID) analyzers (Models 200, 300-HFID, and 300M-HFID; California Analytical; Orange, California). Continuous measurements were recorded every ten seconds. $PM_{2.5}$ (particulate matter with aerodynamic diameter ≤ 2.5 μm) mass was measured gravimetrically with a microbalance (Model MC5; Sartorius; Göttingen, Germany). The $PM_{2.5}$ was sampled isokinetically and collected on polytetrafluoroethylene (PTFE) membrane filters positioned downstream of a $PM_{2.5}$ cyclone (URG, Chapel Hill, NC). Filters were equilibrated at 35% relative humidity and 23°C in an environmental chamber prior to weighing. A particle mobility diameter range of 14.6-661 nm was measured with a SMPS, consisting of an electrostatic classifier and condensation particle counter (Models 3080 and 3010; TSI; Shoreview, Minnesota). UFP emissions over the 14.6-100 nm range were reported on a particle number basis. The SMPS conducted a full scan every 150 seconds, and some short-duration emission events may have been missed. Emissions were quantified using the mass-flow method which requires continuous monitoring of the dilution tunnel air flow and temperature over the WBT measurement period[37].

Controlled laboratory measurements of cookstove emissions sometimes poorly predict field-based emissions[26-28, 40]. Some studies combine data from WBT phases for benchmarking purposes[16] or for comparing intra-study laboratory and field data[28], but these data are based on cookstove usage patterns that may not be reflective of actual field use. To improve the ability to compare laboratory and field measurements, WBT phase-specific emissions data are reported in this study, because each WBT phase simulates, to some extent, different cookstove use. For example, data from the high-power phase of the WBT may be useful for comparison with data from field tests if a cookstove is usually operated at high-power in the field. Thus, cookstove emissions data by WBT phase are likely to be useful in developing international standards. The entire testing database (see the Supporting Information) is provided by the WBT phases for this study. The high-power, cold-start phase is selected for further detailed evaluation below, because (i) emissions tend to be high during this phase, especially for stoves with large thermal mass, and (ii) thermal energy delivered to the cooking pot is not adequately measured in the lower power phase of the WBT. Eventually, a practical method may be developed for accurately measuring cooking energy delivered during low-power so that thermal efficiency can be used as a metric for all phases. Pollutant emissions are analyzed further below.

RESULTS

For each cookstove system, figures and tables showing CO, $PM_{2.5}$, CO_2, THC, and CH_4 mass emission factors—on the basis of time, fuel energy, cooking energy, fuel mass, and WBT cooking tasks (cold-start, hot-start, simmer)—are provided in the Supporting Information. Emissions are reported on an equivalent dry fuel mass basis as defined by the WBT. UFP number emissions, cookstove power, WBT time-to-boil, efficiency, and fuel use are also included. Emission factors may be used to approximate pollutant exposures, to support regional air quality inventories, and for dispersion model input.

Cooking Power and Time-to-Boil Relationship. Figure 1 shows how average cooking power correlates to the time-to-boil for a given pot and volume of water (2L and 5L) and includes data for the cold- and hot-start phases. Cooking power is measured in watts (W) and is defined as the useful cooking energy delivered per unit time, whereas time-to-boil as defined by the WBT is the elapsed time required to boil a specific volume of water. Figure 1 shows that the time-to-boil is a power function of cooking energy and illustrates why the cookstove pot and water volume tested must be appropriate for the stove cooking power. For example, if the pot and volume of water are too large, then the time-to-boil is too long and inconsistent between test replications due to phase change and evaporation[41]. Appropriate use of stove cooking power also produces more consistent task-based results for other WBT parameters such as specific fuel consumption and emissions. Additional test results for fire power (energy released by the fuel per time), cooking power, and time-to-boil are reported in the Supporting Information.

Stove Efficiencies. Modified combustion efficiency (MCE, defined as $CO_2/(CO_2+CO)$ on a molar basis) is considered a reasonable proxy for true combustion efficiency (ratio of energy released by combustion to energy in the fuel)[42]. MCE is equivalent to nominal combustion efficiency (NCE), as used in some literature[9]. Heat transfer efficiency (HTE) refers to the fraction of the heat released by combustion that is used in cooking. Overall thermal efficiency (OTE), the product of MCE and HTE, is the ratio of cooking energy delivered to fuel energy and is an indicator of stove energy efficiency (see the Supporting Information). Figure 2 compares MCE and OTE for the high-power (cold-start) phase of the WBT considering low-moisture fuel; the most efficient stoves are in the upper right corner. Figures 2-5 report test replication error as ± one standard deviation as specified by the WBT. Cookstoves and their performance are often classified[7-34] using fuel type, combustion chamber type, chimney use, heat transfer devices (e.g., pot skirt), draft type (forced or natural convection), fuel feeding or loading method, design, or other characteristics (Table 1). Figures 2-5 present testing results for the individual cookstove systems using the following classification scheme: *3-stone fire, charcoal, forced-draft, natural-draft, and liquid-fuel*. Although instructive for comparing results, we caution that stove classifications can be problematic for standards development (e.g., different benchmarks for stoves with and without chimneys have been proposed[16] but not widely adopted). Classification schemes are non-ideal due to poorly represented and defined stove variations within classes and the inability of novel cookstove technologies to properly fit into what was previously defined. Thus, appropriate standards are best specified based on absolute performance metrics as discussed below.

Cookstoves that achieve both high MCE and OTE show less fuel use and decreased pollutant emission factors. Baseline 3-stone fires (both minimally and carefully tended cases) had approximately 96-97% MCE and approximately 14-15% OTE. Several cookstoves (Sampada, Mayon, StoveTec, Berkeley, Envirofit) with similar MCE as the 3-stone fire show at least two-fold greater OTE. Since these cookstoves consume less fuel, they generally produce lower emissions per given cooking task (as discussed below), but have little or no reductions in emissions per unit fuel. Charcoal stoves show generally low but highly varied MCE due to high CO emissions and nonuniform lump charcoal fuel structure, respectively. Between test replications, airflow differences through the combustion chamber are likely caused by nonuniform charcoal fuel structure. Not all forced-draft stoves exhibit the high MCE they are typically noted for. Forced-draft stoves with fans require electrical energy from household power, rechargeable batteries, or thermoelectric systems (no thermoelectric stove was tested for this study). A natural-draft cookstove with a top-lit up-draft (TLUD) design, see Roth[34], had the highest MCE and OTE, but requires processed, low-moisture, pellet fuel. Assuming unequal variance, the Student's *t*-test (all *p*-values subsequently reported are based on this statistical test) indicated a significant difference ($p \leq 0.024$) between the OTE of the TLUD stove and that of every other stove. Compared with the open-fire base-case, advanced cookstoves (Oorja, Protos, Philips fan, StoveTec TLUD) show improvements in combustion efficiency as indicated by MCE. Relatively minor improvements in combustion efficiency can result in large emissions reductions assuming OTE is maintained. The natural-draft chimney stove (Onil) shows a high MCE but relatively low OTE due to the large thermal mass steel griddle top (termed "plancha" in Latin America). These stoves are used for boiling water but are also used for preparing a variety of foods (e.g., tortillas) and for warmth (space heating). It is noteworthy that the WBT does not apply when stoves are used for purposes other than boiling water. Additional WBT phase-based data for MCE, OTE, and fuel use are provided in the Supporting Information.

Emissions of CO and $PM_{2.5}$. Figure 3 shows the CO and $PM_{2.5}$ emissions per unit cooking energy delivered for low-moisture fuel during the WBT high-power (cold-start) phase. The low emissions stoves reside in the bottom left corner of the figure. The majority of cookstoves emits less CO and $PM_{2.5}$ per unit energy delivered than the 3-stone fire base-case. Two forced-draft stoves (Philips fan, Oorja) and the TLUD-type stove had notably low emissions. A significant difference ($p \leq 0.018$) was observed for the TLUD stove CO emissions compared with every other stove. Charcoal stoves emit high CO levels during all three WBT phases and high PM emissions during the cold-start phase due to the charcoal ignition process. After ignition, charcoal stoves can produce high levels of hazardous, odorless CO with much less warning in the form of irritating smoke compared to wood stoves and thus should be tested and used in well ventilated areas only. An intermittent problem with the liquid-fuel stove burner caused high PM emissions variability and possibly higher than expected PM emissions.

Compared with the high-power level, the two rocket stoves operated at medium power show higher MCE and OTE (Figure 2) and lower CO and $PM_{2.5}$ mass emissions when normalized to cooking energy delivered (Figure 3). The difference in MCE for the two power levels was significant ($p = 0.0005$) for the StoveTec stove. These rocket stoves can thus achieve lower emissions for a given cooking task at less than maximum power. This comparison demonstrates the value of evaluating cookstoves at an additional power level for developing international standards. United States Environmental Protection Agency (USEPA) certification testing requires four power levels for residential, wood-fueled, heating stoves[43]. An additional benefit of stove testing at multiple power levels is the ability to better correlate laboratory results with field results[42].

Figure 4 shows the CO and $PM_{2.5}$ emissions per liter of water simmered per hour for low-moisture fuel considering the WBT low-power phase. The majority of cookstoves emit less CO and $PM_{2.5}$ per unit volume of water per time than the 3-stone fire base-case. A forced-draft stove (Philips fan) had notably low emissions. Charcoal stoves emit lower PM levels during the WBT low-power phase (Figure 4) than during the high-power cold-start phase (Figure 3). Again, an intermittent problem with the liquid-fuel stove burner caused high PM emissions variability and possibly higher than expected PM emissions.

UFP Emissions. Figure 5 shows the UFP number and $PM_{2.5}$ mass emissions per cooking energy delivered for low-moisture fuel and the high-power (cold-start) phase. UFPs are of interest because they can penetrate deep into the airways of the human respiratory tract to the alveoli, where they may cause adverse biological effects[44]. Presently, there are no USEPA standards or WHO guidelines related to UFPs, although European Union vehicle emissions legislation does consider UFPs. The majority of cookstoves tested show lower UFP emissions compared with the 3-stone fire. Intermittent malfunction of a fan speed controller likely produced highly variable UFP emissions for the Oorja stove. A natural-draft TLUD stove shows the lowest mean UFP and $PM_{2.5}$ mass emissions. The UFP emissions of the TLUD stove were significantly lower ($p = 0.0007$) than those of the forced-draft Philips fan stove. Forced-draft stoves emit relatively less $PM_{2.5}$ mass but show an increase in UFP numbers[45]; in this case, gas phase nucleation may be occurring in an environment where fewer accumulation mode particles produce less surface area for condensation and growth[46].

DISCUSSION

The extensive testing and metrics analysis performed as part of this study allows for further insight critical to advancing the development of realistic international cookstove testing and emissions standards. This study considers multiple cookstove performance metrics (see the Supporting Information) and recommends potential metrics for future cookstove standards. Pollutant emissions per cooking energy delivered[47] (in units of g $MJ_{delivered}^{-1}$) and OTE are recommended measures for the high-power WBT phases because they are based on the fundamental desired output – cooking energy – that enables valid comparisons between all stoves and fuels[9]. For high-power WBT phases, the cooking energy delivered is determined by (i) the sensible heat that raises the pot water temperature and (ii) the latent heat that produces steam. A relatively small quantity of energy is unaccounted for (as loss from the pot), but OTE is measured accurately. This is not so for the low-power WBT phase, despite the previous use of the OTE metric, because (i) relatively constant water temperatures result in limited or no measured sensible heat and (ii) highly variable steam production produces variation in measured latent heat. Furthermore, the unaccounted energy can be substantial relative to the latent heat. Thus, alternative metrics are recommended for the low-power phase due to the lack of a method for measuring energy delivered during low-power. Development of a practical method for accurately measuring cooking energy delivered during the low-power phase would enable the use of the same metrics for all WBT phases. Until such a method is developed, specific energy consumption (SEC – in units of MJ L^{-1} h^{-1}) and specific emission rate (SER– in units of g L^{-1} h^{-1}) are recommended as standard measures for the WBT low-power phase. SEC is energy utilization and SER is emissions per liter of water maintained at the WBT-specified temperature per unit time. The importance of documenting cooking power for the purpose of meeting end-user needs is noted. Time-to-boil may also be reported if the pot type and water volume are specified, but time-to boil can be a misleading indicator due to the nonlinear correlation with cooking power, as illustrated in Figure 1 and discussed above.

Baldwin[47] proposed “cooking process efficiency” as an ultimate metric that may include, for example, the use of a pressure cooker to improve process efficiency. Cooking process efficiency also includes “control efficiency” which indicates the ability to provide “…only as much heat as needed to cook the food…”[47]. The WBT protocol specifies a “turn-down ratio” metric–the ratio of high-power to low-power—indicating the extent to which a stove can be controlled. However, this WBT standard measure is limited in that it does not account for the wide range of power offered by some stoves nor does it indicate a stove’s ability to respond rapidly to power level adjustments—issues that a revised WBT may consider.

This study attempted to test realistic cookstove systems to improve relevance to field conditions, as discussed above, but further investigation is needed. Laboratory testing provides a cost-effective means of evaluating cookstoves while controlling variables that are difficult or impossible to control in the field. Despite its advantages controlled laboratory testing cannot fully duplicate field testing, but should emulate field conditions to the greatest extent possible[28]. Expanded field research is needed to provide critical information on actual use conditions that cannot be duplicated from controlled tests. Thus, there is a need for future emphasis on coordination between controlled and field testing. Development of test protocols is needed for stoves performing tasks substantially different than boiling water, such as the griddle stoves discussed above.

Despite a lack of correlation with emissions of some other pollutants that also affect health[48] and climate[40], CO and PM emission measurements continue to be widely acquired for cookstoves due to their relative simplicity and availability.

CO and PM measurements are likely to be required as part of any standardization process. The USEPA uses gravimetric PM measurement for its National Ambient Air Quality Standards and for its certification programs, which include measuring emissions from residential, wood-fueled, heating stoves sold in the U.S.[43]. The gravimetric method is generally considered more accurate and reliable[49] compared with relatively low-cost optical methods typically used in the developing world for PM measurement. Nevertheless, light-scattering instruments can be useful for obtaining real-time emissions data needed for improving cookstove designs and for better understanding human exposures to air pollutants. A relatively low-cost gravimetric measurement method for PM is needed to enable widespread testing capacity to evaluate stoves against international standards. Emissions of other important organic and inorganic pollutants also require characterization using newly-developed, low-cost, accurate, and rapid analytical methods.

Cookstoves with chimneys can produce fugitive emissions from the stove body into the indoor environment. Despite the difficulty of mimicking fugitive emissions in laboratories, future evaluations of chimney stoves should consider both indoor fugitive and total emissions consistent with the present study. Indoor emissions have a greater effect on household air quality and human health, while total emissions have a greater effect on outdoor air quality and climate. Oddly, more performance data are currently available for newer biomass stoves than for the traditional and modern stoves dominating usage worldwide. To better align expectation levels with realized stove improvements, future performance evaluations must include "best-case" liquid- and gas-fueled stoves[50] as well as more types of "worst-case" traditional solid-fuel stoves, based on the same absolute performance metrics.

Outlook. The WBT protocol is currently being revised, and work is needed to finalize the revision[37]. With the 2010 launch of the Global Alliance for Clean Cookstoves (GACC), "a major global cookstove renaissance"[51] offers the opportunity to build on the foundational work of the Partnership for Clean Indoor Air (PCIA) and partner organizations around the world – see background in the Supporting Information. The GACC Standards and Testing Working Group recognized the need to build on prior work to improve controlled and field evaluation of household cookstoves and the need to build capacity in the developing world for evaluating and improving cookstoves[52]. Stakeholders present at the 2011 PCIA Forum in Lima, Peru, including some members of the Standards and Testing Working Group, drafted the "Lima Consensus"[53], an agreement to establish an interim rating system for the evaluation of cookstove models "that reflects the varying tiers of performance in the areas of fuel efficiency, indoor air quality, $PM_{2.5}$ and CO emissions, and safety." A rating system was proposed to be stove- and fuel-neutral, simply rating stove/fuels by a number of criteria that will better communicate the performance of existing stove models and drive innovation to improve stove performance. Building on the Lima Consensus, an International Organization for Standardization (ISO) International Workshop Agreement (IWA) was finalized and unanimously affirmed by more than 80 stakeholders present at The Hague, Netherlands on February 28-29, 2012. Recommendations from this work were adopted in the IWA entitled *Guidelines for Evaluating Cookstove Performance*[54]. This agreement is an important step toward developing the methods and metrics to be used in international cookstove standards. Results from this study are mapped to Tiers defined in the IWA (see the

Supporting Information). Emissions standards will also depend on assessing what is needed to protect health. This is currently being addressed under the World Health Organization's Air Quality Guidelines program[55]. Other important cookstove characteristics that were not evaluated in this study include safety, durability, cost, controllability, and user acceptability. Evaluation methods and metrics for these other important characteristics need to be further developed.

Supporting Information. Additional information is provided on the cookstoves, fuels, pots, facility, and test protocol used in this study, as well as supplemental technical discussion. A database is provided for cookstove systems tested, which includes results for fuel moisture, fuel energy, cookstove power, WBT time-to-boil, efficiency, fuel use, and emissions of CO, $PM_{2.5}$, CO_2, THC, CH_4, and UFPs. This information is available free of charge via the Internet at http://pubs.acs.org/ .

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REFERENCES

(1) *Global Health Risks: Mortality and Burden of Disease Attributable to Selected Major Risks.* World Health Organization: Geneva, Switzerland, 2009.

(2) *Global Forest Resources Assessment 2010.* Food and Agriculture Organization of the United Nations: Rome, Italy, 2010.

(3) *Special Report on Renewable Energy Sources and Climate Change Mitigation.* Intergovernmental Panel on Climate Change: Geneva, Switzerland, 2011.

(4) Smith, K. R. Health, energy, and greenhouse-gas impacts of biomass combustion in household stoves. *Energy for Sustainable Development*, **1994**, *1* (4), 23-29.

(5) 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 randomised controlled trial. *The Lancet* **2011**, *378*, 1717-1726.

(6) *Household Cookstoves, Environment, Health, and Climate Change: A New Look at an Old Problem.* The World Bank: Washington, DC, 2011.

(7) Ballard-Tremeer, G.; Jawurek, H. H. Comparison of five rural, wood-burning cooking devices: efficiencies and emissions. *Biomass & Bioenergy* **1996**, *11*(5), 419-430.

(8) Oanh, N. T. K.; Reutergårdh, L. B. R.; Dung, N. T. Emission of polycyclic aromatic hydrocarbons and particulate matter from domestic combustion of selected fuels. *Environ. Sci. Technol.* **1999**, *33*(16), 2703-2709.

(9) Smith, K. R.; Uma, R.; Kishore, V. V. N.; Lata, K.; Joshi, V.; Zhang, J.; Rasmussen, R. A.; Khalil, M. A. K. *Greenhouse Gases from Small-scale Combustion Devices in Developing Countries.* United States Environmental Protection Agency: Washington D.C., EPA/600/R-00/052, 2000.

(10) Bhattacharya, S. C.; Albina, D. O.; Salam, P. A. Emission factors of wood and charcoal-fired cookstoves. *Biomass & Bioenergy* **2002** *23*, 453-469.

(11) Oanh, N. T. K.; Nghiem, L. H.; Phyu, Y. L. Emission of polycyclic aromatic hydrocarbons, toxicity, and mutagenicity from domestic cooking using sawdust briquettes, wood, and kerosene. *Environ. Sci. Technol.* **2002**, *36*(5), 833-839.

(12) Oanh, N. T. K.; Albina, D. O.; Ping, L.; Wang, X. Emission of particulate matter and polycyclic aromatic hydrocarbons from select cookstove-fuel systems in Asia. *Biomass Bioenergy* **2005**, *28*, 579-590.

(13) 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 stoves. *Energy for Sustainable Development* **2008**, *12*, 5-14.

(14) Yuntenwi, E. A. T.; MacCarty, N.; Still, D.; Jürgen, E. Laboratory study of the effects of moisture content on heat transfer and combustion efficiency of three biomass cook stoves. *Energy for Sustainable Development* **2008**, *12*, 42-57.

(15) Jetter, J. J.; Kariher, P. Solid-fuel household cook stoves: Characterization of performance and emissions. *Biomass & Bioenergy* **2009**, *33*, 294-305.

(16) MacCarty, N.; Still, D.; Ogle, D. Fuel use and emissions performance of fifty cooking stoves in the laboratory and related benchmarks of performance. *Energy for Sustainable Development* **2010**, *14*, 161-171.

(17) Zhang, J.; Smith, K. R.; Uma, R.; Ma, Y.; Kishore, V. V. N.; Lata, K. Carbon monoxide from cookstoves in developing countries: 1.Emission factors. *Chemosphere Global Change Sci.* **1999**, *1*, 353-366.

(18) Zhang, J.; Smith, K. R. Emissions of carbonyl compounds from various cookstoves in China. *Environ. Sci. Technol.* **1999**, *33*(14), 2311-2320.

(19) Zhang, J.; Smith, K. R.; Ma, Y.; Ye, S.; Weng, X.; Jiang, F. Greenhouse gases and other pollutants from household stoves in China: A database for emission factors. *Atmos. Environ.* **2000**, *34*(26), 4537-4549.

(20) Tsai, S. M.; Zhang, J.; Smith, K. R.; Ma, Y.; Rasmussen, R. A.; Khalil, M. A. K. Characterization of non-methane hydrocarbons emitted from various cookstoves used in China. *Environ. Sci. Technol.* **2003**, *37*, 2869-2877.

(21) Gupta, S.; Saksena, S.; Shankar, V. R.; Joshi, V. Emission factors and thermal efficiencies of cooking biofuels from five countries. *Biomass Bioenergy* **1998**, *14*, 547-559.

(22) McCracken, J. P.; Smith, K. R. Emissions and efficiency of improved woodburning cookstoves in highland Guatemala. *Environment International*, **1998**, *24* (7), 739-747.

(23) Shen, G.; Yang, Y.; Wang, W.; Tao, S.; Zhu, C.; Min, Y.; Xue, M.; Ding, J.; Wang, B.; Wang, R.; Shen, H.; Li, W.; Wang, X.; Russell, A. G. Emission factors of particulate matter and elemental carbon for crop residues and coals burned in typical household stoves in China. *Environ. Sci. Technol.*, **2010**, *44*, 7157-7162.

(24) Shen, G; Wang, W.; Yang, Y.; Ding, J.; Xue, M.; Min, Y.; Zhu, C.; Shen, H.; Li, W.; Wang, B.; Wang, R.; Wang, X.; Tao, S.; Russell, A. G. Emissions of PAHs from indoor crop residue burning in a typical rural stove: Emission factors, size distribution, and gas – particle partitioning. *Environ. Sci. Technol.*, **2011**, *45*, 1206–1212.

(25) Shen, G.; Tao, S.; Wang, W.; Yang, Y.; Ding, J.; Xue, M.; Min, Y.; Zhu, C.; Shen, H.; Li, W.; Wang, B.; Wang, R.; Wang, W.; Wang, X.; Russell, A. G. Emission of oxygenated polycyclic aromatic hydrocarbons from indoor solid fuel combustion. *Environ. Sci. Technol.*, **2011**, *45*, 3459-3465.

(26) Bailis, R.; Berrueta, V.; Chengappa, C.; Dutta, K.; Edwards, R.; Masera, O.; Still, D.; Smith, K. Performance testing for monitoring improved biomass stove interventions: experiences of the Household Energy and Health Project. *Energy for Sustainable Development* **2007**, *11*, 57-70.

(27) Johnson, M.; Edwards, R.; Frenk, C. A.; Masera, O. In-field greenhouse gas emissions from cookstoves in rural Mexican households. *Atmospheric Environment* **2008**, *42*, 1206-1222.

(28) 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. *Atmospheric Environment* **2009**, *43*, 1170-1181.

(29) Smith, K. R.; Khalil, M. A. K.; Rasmussen, R. A.; Thorneloe, S. A.; Manegdeg, F.; Apte, M. Greenhouse gases from biomass and fossil fuel stoves in developing countries: A Manila pilot study. *Chemosphere* **1993**, *26*, 479-505.

(30) Venkataraman, C.; Rao, G. U. M. Emission factors of carbon monoxide and size-resolved aerosols from biofuel combustion. *Environ. Sci. Technol.* **2001**, *35*(10), 2100-2107.

(31) Venkataraman, C.; Negi, G.; Sardar, S. B.; Rastogi, R. Size distributions of polycyclic aromatic hydrocarbons in aerosol emissions from biofuel combustion. *J. Aerosol Science* **2002**, *33*(3), 503-518.

(32) Chen, Y.; Bi, X.; Mai, B.; Sheng, G.; Fu, J. Emission characterization of particulate/gaseous phases and size association for polycyclic aromatic hydrocarbons from residential coal combustion. *Fuel* **2004**, *83*, 781-790.

(33) Chen, Y.; Sheng, G.; Bi, X.; Feng, Y.; Mai, B.; Fu, J. Emission factors for carbonaceous particles and polycyclic aromatic hydrocarbons from residential coal combustion in China. *Environ. Sci. Technol.* **2005**, *39*(6), 1861-1867.

(34) Roth, C. *Micro-Gasification: Cooking with Gas from Biomass*. Deutsche Gesellschaft für Internationale Zusammenarbeit (GIZ) GmbH: Eschborn, Germany, 2011.

(35) *ASTM D4442-07 Standard Test Methods for Direct Moisture Content Measurement of Wood and Wood-Base Materials.* ASTM International: West Conshohocken, Pennsylvania, 2007.

(36) *ASTM D5865-10 Standard Test Method for Gross Calorific Value of Coal and Coke*. ASTM International: West Conshohocken, Pennsylvania, 2010.

(37) *Water Boiling Test (WBT) Version 4.* Partnership for Clean Indoor Air: Washington, DC., 2010. http://www.pciaonline.org/testing

(38) Hildemann, L. M.; Cass, G. R.; Markowski, G. R. A dilution stack sampler for collection of organic aerosol emissions: Design, characterization and field tests. *Aerosol Sci. Technol.* **1989**, *10*, 193-204.

(39) Hays, M. D.; Geron, C. D.; Linna, K. J.; Smith, N. D.; Schauer, J. J. Speciation of gas-phase and fine particulate emissions from burning of foliar fuels. *Environmental Science and Technology* **2002**, *36* (11), 2281-2295.

(40) Roden, C. A.; Bond, T. C.; Conway, S.; Benjamin, A.; Pinel, O. Emission factors and real-time optical properties of particles emitted from traditional wood burning cookstoves. *Environmental Science and Technology* **2006**, *40*, 6750-6757.

(41) L'Orange, C.; DeFoort, M.; Willson, B. Influence of testing parameters on biomass stove performance and development of an improved testing protocol . *Energy for Sustainable Development* **2012**, *16*, 3-12.

(42) Johnson, M.; Edwards, R.; Berrueta, V.; Masera, O. New approaches to performance testing of improved cookstoves. *Environmental Science and Technology* **2010**, *44*, 368-374.

(43) *Code of Federal Regulations (CFR), Title 40, Part 60, Subpart AAA - Standards of Performance for New Residential Wood Heaters.* United States Environmental Protection Agency: Washington, D.C., 53 Federal Register 5873, Feb. 26, 1988.

(44) Valavanidis, A.; Fiotakis, K.; Vlachogianni, T. Airborne particulate matter and human health: Toxicological assessment and importance of size and composition of particles for oxidative damage and carcinogenic mechanisms. *Journal of Environmental Science and Health* **2008**, *26* (C), 339–362.

(45) Hawley, B.; Volckens, J. Proinflammatory effects of cookstove emissions on human bronchial epithelial cells. *Indoor Air* **2012**, in press.

(46) Khalek, I. A.; Kittelson, D. B.; Brear, F. Nanoparticle growth during dilution and cooling of diesel exhaust: Experimental investigation and theoretical assessment. SAE International: Warrendale, PA. **2000**. DOI 10.4271/2000-01-0515.

(47) Baldwin, S. F. *Biomass Stoves.* Volunteers in Technical Assistance: Arlington, Virginia. 1987.

(48) Sahu, M.; Peipert, J.; Singhal, V.; Gautam, N. Y.; Biswas, P. Evaluation of mass and surface area concentration of particle emissions and development of emissions indices for cookstoves in rural India. *Environ. Sci. Technol.* **2011**, *45*, 2428-2434; DOI 10.1021/es1029415.

(49) Hinds, W. C. *Aerosol Technology*; John Wiley & Sons, Inc.: New York, USA, 1999; p. 218.

(50) Smith, K. R. “Cooking with Gas.” *Energy for Sustainable Development*. **2011**, *15*, 115-116.

(51) Smith, K. R. What’s cooking? A brief update. *Energy for Sustainable Development* **2010**, *14*, 251-252.

(52) *GACC Working Group Early Action Items*. Global Alliance for Clean Cookstoves: Washington, DC, 2011. http://cleancookstoves.org/wp-content/uploads/2011/06/Early-Action-Item-Compilation-FINAL.pdf

(53) *The “Lima Consensus.”* Partnership for Clean Indoor Air: Washington, D.C., 2011. http://www.pciaonline.org/testing/lima-consensus

(54) *International Workshop Agreement 11:2012, Guidelines for Evaluating Cookstove Performance.* International Organization for Standardization: Geneva, Switzerland, 2012.

(55) *Indoor Air Pollution and Health.* World Health Organization: Geneva, Switzerland, 2011. http://www.who.int/mediacentre/factsheets/fs292/en/

Table 1. Fuels and Cookstoves Tested

Fuel	Tested with Low-/ High-moisture fuel	Cookstove	Cooking pot, water volume (liters)	Combustion chamber material	Chimney	Pot skirt	Forced-draft	Batch fuel	Rocket-type	Gasifier-type	Supporting Info I.D.
Wood	L/H	3-stone, carefully tended	Standard, 5	None							A
	L	3-stone, minimally tended	Standard, 5	None				●			A
	L/H	Berkeley Darfur	Rnd-btm. Alum., 5	Metal		●					B
	L/H	Envirofit G-3300	Standard, 5	Metal		●			●		C
	L	Onil	Standard, 5	Ceramic	●				●		D
	L/H	Philips HD4008	Standard, 5	Metal						●	E
	L/H	Philips HD4012	Standard, 5	Ceramic			●			●	F
	L/H	Sampada	Standard, 5	Metal						●	G
	L/H	StoveTec GreenFire	Standard, 5	Ceramic		●			●		H
	L/H	Upesi Portable	Rnd-btm. Alum., 5	Ceramic							I
Charcoal	L/H	GERES	Standard, 5	Ceramic				●			J
	L/H	Gyapa	Standard, 5	Ceramic				●			K
	L/H	Jiko, ceramic	Standard, 2	Ceramic				●			L
	L/H	Jiko, metal	Standard, 2	Metal				●			M
	L/H	KCJ Standard	Standard, 5	Ceramic				●			N
	L/H	Kenya Uhai	Standard, 5	Ceramic				●			O
	L/H	StoveTec prototype	"Superpot", 5	Ceramic		●		●	●		P
Rice hulls	L/H	Belonio Rice Husk Gasifier	Standard, 2	Metal			●	●		●	Q
	L/H	Mayon Turbo	Standard, 5	Metal						●	R
Pellets, Oorja	L/H	Oorja	Standard, 2	Ceramic			●	●		●	S
Pellets, wood	L	StoveTec TLUD prototype	Standard, 2	Metal		●		●		●	T
Corn cobs	L/H	Jinqilin CKQ-80I	Standard, 5	Metal	●		●			●	U
Plant oil	n/a	Protos	Standard, 2	n/a				●			V

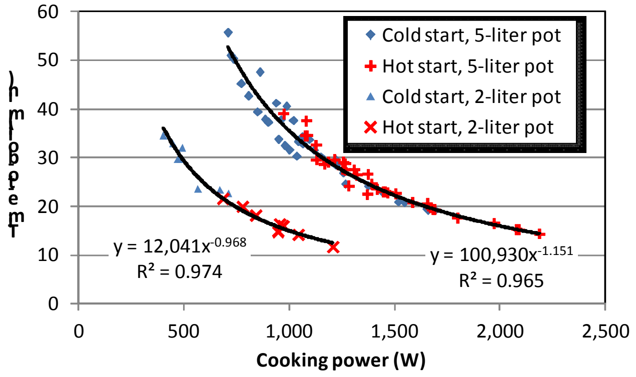

Figure 1. Time-to-boil versus cooking power for all cookstove systems evaluated

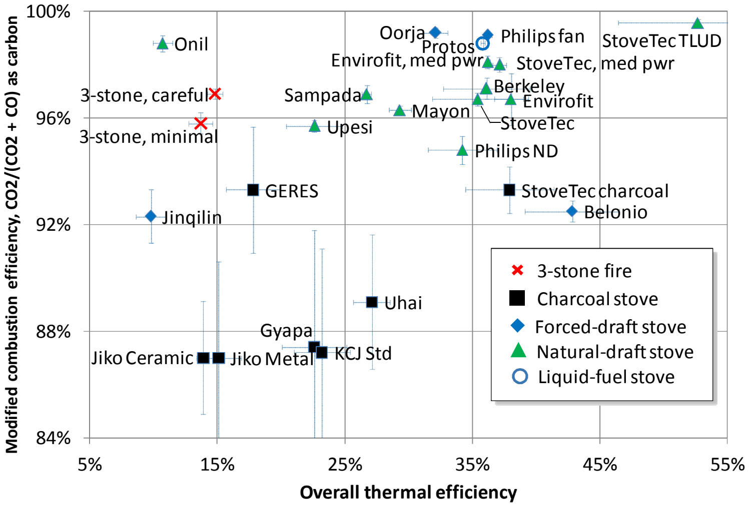

Figure 2. MCE versus OTE for low-moisture fuel during the high-power (cold-start) phase of the WBT.

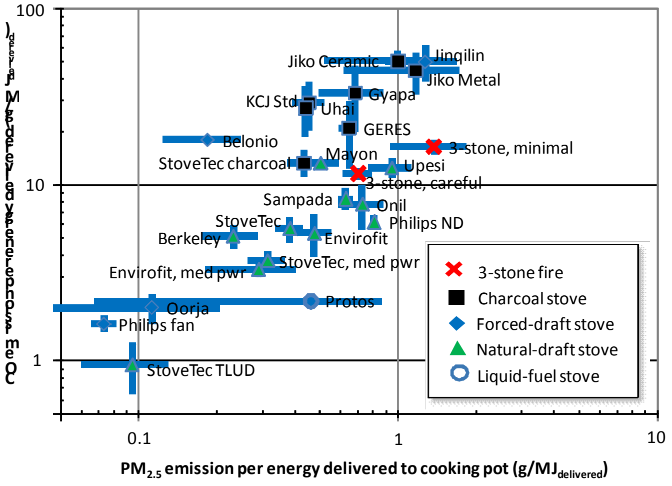

Figure 3. CO compared to $PM_{2.5}$ emissions per energy delivered to the cooking pot for low-moisture fuel during the high-power (cold-start) phase of the WBT.

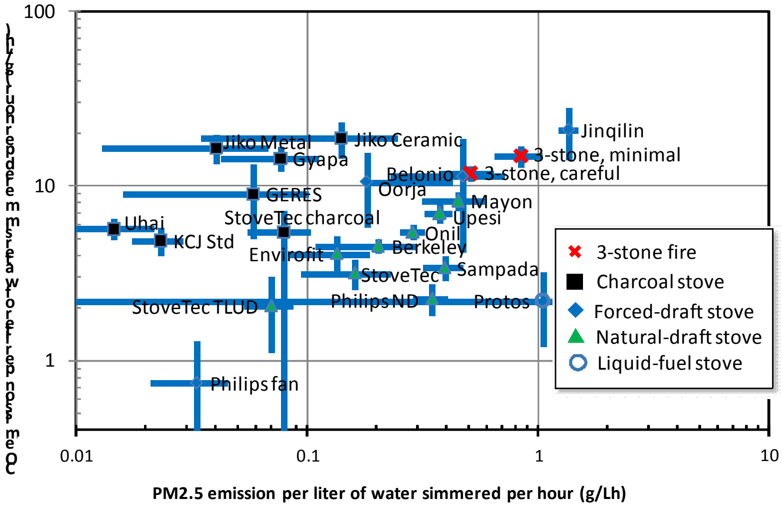

Figure 4. CO compared to $PM_{2.5}$ emissions per liter of water simmered per hour for low-moisture fuel during the low-power phase of the WBT.

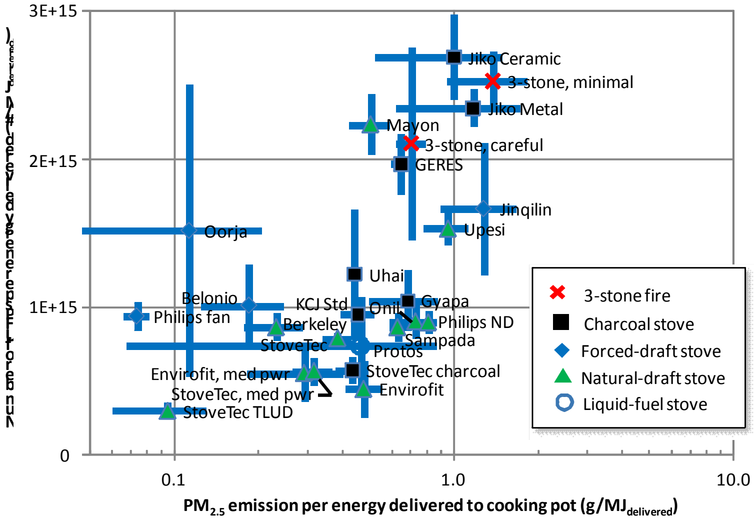

Figure 5. Number of UFPs compared to $PM_{2.5}$ emissions per energy delivered to the cooking pot for low-moisture fuel during the high-power (cold-start) phase of the WBT.

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