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Efficiencies and pollutant emissions from forced-draft biomass-pellet semi-gasifier stoves: Comparison of International and Chinese water boiling test protocols

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ABSTRACT

Biomass fuels are widely combusted in rural China, producing numerous air pollutants with great adverse impacts on human health. Some improved cookstoves and pellet fuels have been promoted. To evaluate the performance of pellet-gasifier stoves, efficiencies and pollutant emissions were measured following International and Chinese water boiling tests (WBTs). Compared with traditional stoves and unprocessed biomass fuels, increased efficiencies and lower emissions of pollutants including carbon monoxide (CO), particulate matter (PM), parent and derivative polycyclic aromatic hydrocarbons (PAHs) were revealed for pellet-gasifier stoves. However, the calculated emission rates (ERs) of CO and $PM_{2.5}$ cannot meet the ER targets recently suggested by WHO indoor air quality guidelines (IAQGs). Better control of air mixing ratio and gross flow rates of primary and secondary air supply greatly reduced emissions and increased efficiencies. Differences among testing protocols are the key factors affecting the evaluation of stove performance. With longer burning duration and higher power, the Chinese WBT had statistically higher efficiencies, gas temperature, and lower pollutant emissions ($p < 0.10$) compared to those obtained through the International WBT. Statistically significant differences between the two protocols indicate the need for further efforts in emission tests and methodology development before the release of a well-accepted international testing protocol.

Introduction

Globally, over 2.6 billion people are still relying on traditional biomass fuels for household cooking activities (International Energy Agency, IEA, 2013). Incomplete burning of traditional fuels usually produces large amounts of air pollutants, including CO, PM, black carbon (BC), and organics like PAHs, and subsequently leads to severe household air pollution, adverse impacts on human health, and local and regional climate change (Reid et al., 2012; Smith, 2013a; WHO, 2009; Rao et al., 2013). Residential fuel combustion is one major source of many incomplete combustion products, especially in developing countries. Household air pollution has been recognized as one of the top environmental risk factors affecting human health globally and results in approximately four million premature deaths annually (Lim et al., 2013; Zhang and Smith, 2007; Smith et al., 2013b, 2014).

Traditional stoves were often lower in heating transfer efficiency (HTE) and thermal efficiency, had a long time duration for cooking, consumed a large amount of fuels, and produced high pollutant emissions. Consequently, notable adverse impacts on air quality and human health are yielded (Edwards et al., 2004; Jetter et al., 2012; Clark et al., 2013; Shen et al., 2015a). Efforts have been made to increase HTE and/or thermal efficiency in the stoves' performance, so as to reduce fuel consumption and lower air pollution (Smith et al., 2000; Jetter et al., 2012; Dutt and Ravindranath, 1993; Shen et al., 2015b; Kshirsagar and Kalamkar, 2014). The experience in China showed that the development of stoves experienced four stages (Shen et al., 2015b). Improved stoves were promoted and benefited air quality and human health from the 1980s (some simple improved stoves with

Abbreviations: BC, black carbon; CCT, controlled cooking test; CO, carbon monoxide; EC, elemental carbon; EF, emission factor; ER, emission rate; GFF, glass fiber filter; HTE, heat transfer efficiency; IAQ, indoor air quality; IAQG, indoor air quality guideline; KPT, kitchen performance test; LHV, lower heating value; MCBM, Monte Carlo box model; MCE, modified combustion efficiency; MDL, method detection limit; OC, organic carbon; OTE, overall thermal efficiency; PAH, polycyclic aromatic hydrocarbon; PM, particulate matter; PUF, polyurethane foam plug; QFF, quartz fiber filter; VM, volatile matter; WBT, water boiling test.

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ventilation, grates, and chimney) when the National Improved Stove Program was initiated (Shen et al., 2015b; Smith et al., 1993a). Currently, after the fast research and development of stoves in China, some high-efficiency clean stoves like gasifier stoves with primary and secondary air supply, and forced-draft stoves are available, which are expected to be able to lower pollutant emissions and improve air quality after an effective intervention program.

Evaluating efficiency and emission performance is a good way to compare one fuel–stove combination with another (Jetter et al., 2012). In addition to fuel and stove properties, the burn cycle protocols and other factors like sampling and laboratory analysis can affect the results of emission and efficiency greatly. According to existing standards and guidelines, laboratory-simulated emission measurements can repeat the burning processes, and thus have been widely used in the evaluation and comparison of performance among different fuel–stove combinations. Though the WBT is commonly utilized in laboratory emission measurement, the detailed procedure varies greatly in various protocols (Makonese et al., 2011; Arora et al., 2014). For example, the International WBT is somewhat different from the one (the Chinese WBT) commonly used in China in time control, water temperature, and parameter calculation and description (Water Boiling Test, WBT Version 4.1.2, 2009; Chinese Water Boiling Test, WBT, 2008), as we present in the following method section.

In this study, three gasifier stoves burning pellets were tested for efficiencies, emission factors (EFs) and ERs of CO, PM, elemental carbon (EC), organic carbon (OC), and PAHs in a laboratory using both International and Chinese WBTs methods. The differences among three pellet-gasifier stoves and between the two WBTs are compared and discussed. It is expected that the results will provide important data relevant to clean stove intervention programs and contribute to the development of an international standard test protocol in the future.

Experimental

Fuels and stoves

Three different models of pellet-gasifier stoves sold in some rural areas of China were tested in this study. All of them have primary and secondary air supply devices controlled through a fan. By turning the dials on front of the stoves, the flow rate of primary and secondary air can be adjusted. It is noted that some intervention programs are promoted in rural China, and the stoves in the present study are under strong consideration in these intervention programs (Carter et al., 2014). The photos and detailed manufacturing information are shown in Table 1 and Fig. 1. Stove 1 was purchased from the local market in Shanxi, Northern China, and primary and secondary air supply can be controlled separately. Stove 2 was from Hunan in Southern China. With only one dial in front, it can control the gross air supply fan power; however, the ratio between primary and secondary air is preset by the manufacturer and cannot be altered separately by the user. Stove 3 was from Henan in central China and it adjusts the burning conditions by varying the ratio of primary and secondary air supply under a stable gross air supply. The same batch of pellets made with cornstalk with a small amount of cow dung (~9:1), was used in each stove. A small amount of dry high-resin pinewood (approximately 100 g) was used for initial lighting. The measured carbon content, nitrogen content, hydrogen content, oxygen content (by difference), volatile matter (VM), moisture (wet basis), and lower heating value (LHV) of the pellet were 42%, 1.44%, 6.55%, 55.23%, 65.34%, 14%, and 17.0 MJ/kg, respectively. The ash content was around 9.4%.

Table 1
Information on the three Chinese pellet-gasifier stoves tested in this study

Stove number	Stove model	Production year	Manufacturer	Location
Stove 1	CKQ-80	2009	Jinqilin	Shanxi, China
Stove 2	CLKB 2.5-IY	2010	Xunda	Hunan, China
Stove 3	HLJF-CS 3.5	2011	Heluo	Henan, China

Water boiling tests

The International and Chinese WBT protocols are different in the operation procedure and calculation. Three test phases including cold start, hot start, and simmer phases are tested in the International WBT protocol. The cold start phase starts from the fuel lighting by heating a pot of water (5 L) from the ambient temperature to the boiling point. When the cold start phase is completed, the remaining fuels are weighed. The hot start phase follows with the stove at the same operating procedure and heating another pot of water from ambient to boiling temperature. The simmering phase maintains a measured amount of water at just below the boiling point for 45 min (Water Boiling Test, WBT Version 4.1.2, 2009). In the present test, the simmering phase was not tested as it is seldom used in real practice in China. The pot is not covered during the whole test. As previous studies found that the differences in pollutant emissions between the cold start and hot start of the International WBT was small for stoves with relatively small thermal mass, an averaged value was calculated representing a value for high power performance, as specified by the International Standard Workshop Agreement. tiered stove rating framework (Carter et al., 2014; Water Boiling Test, WBT Version 4.1.2, 2009; International Workshop Agreement, IWA, 2012).

In the Chinese WBT protocol, there is only one test phase (Chinese Water Boiling Test, WBT, 2008). Once fuel is ignited, the pot with 5 L of water and lid is put onto the stove, and the test starts. When the water temperature reaches the boiling point, the pot cover is removed. But remaining fuels are left in the stove chamber and burned. The test ends when the water temperature decreases to 5 °C below the boiling point. The schematic diagram showing the water temperature over time for these two WBTs is provided in Fig. 2.

Calculation

In both Chinese and International WBT protocols, water mass is preweighed, and the mass of water evaporated is measured. Water temperature is measured continuously throughout the test. The initial water temperature, water boiling temperature, and test duration are recorded. These parameters are used to calculate the performance indicators, including thermal efficiency and pollutant EFs.

Overall thermal efficiency (OTE) is a measure of the ratio of useful energy delivered (to the water in the pot) to the fuel energy from complete combustion. The useful energy delivered includes the energy for both water heating and water evaporation. The calculation of OTE in both International and Chinese WBT protocols is the same, using the following equation:

$$OTE = \frac{\Delta E_{H_2O,Heat} + \Delta E_{H_2O,evap}}{E_{\text{released},c}}$$

$\Delta E_{H_2O,heat}$: Calorific heat transferred to water in the pot which was heated from room temperature to boiling point.
$\Delta E_{H_2O,evap}$: Calorific heat transferred to the water in the pot to evaporate.
$E_{released,c}$: Calorific heat delivered by the equivalent dry fuel consumed.

The modified combustion efficiency (MCE), defined as $CO_2/(CO + CO_2)$ (molar basis), is a reasonable proxy for efficiency and also the percentage of the chemical energy in the fuel that is actually released. It indicates how well fuel is burned. HTE is the ratio of energy delivered to the pot versus the total heat energy released from the fuel burning. However, in most circumstances, it is hard to determine HTE. It was

Fig. 1. The photos for stove 1 (left), stove 2 (middle), and stove 3 (right)

also not measured directly in this study. Thus, it was calculated by MCE and OTE using the equation—HTE = OTE/MCE (Smith et al., 1993b, 2000).

To illustrate the pollutant emissions of International and Chinese WBTs, EFs and ERs were used. The carbon mass balance method, which assumed that the total carbon emitted from fuel combustion is in the form of gaseous phase (CO, CO_2, and total hydrocarbons) and particulate carbon fractions, was used to calculate EFs (Shen et al., 2012a,b; Zhang et al., 2000). ERs, in measuring unit of pollutant mass emitted per time, were calculated from pollutant EFs, fuel consumption amount and combustion test duration.

In addition, the ERs of CO and $PM_{2.5}$ from these pellet-gasifier stoves were assessed in terms of IAQ through the method described in the

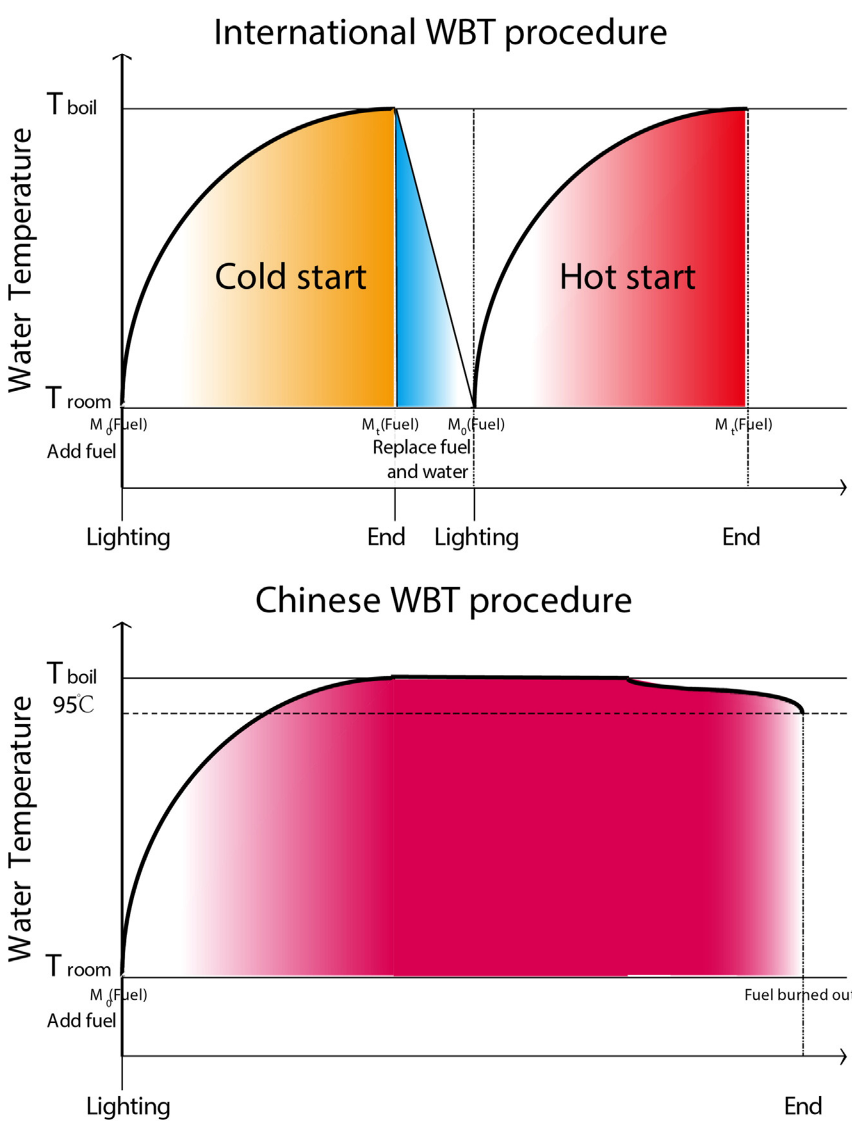

Fig. 2. The schematic diagrams of International and Chinese WBTs

WHO Guidelines, which links the emissions of household energy devices and IAQGs (Johnson et al., 2014). Briefly, a Monte Carlo box model (MCBM) is used to calculate the indoor air concentrations of CO and $PM_{2.5}$ based on some parameters, including ER, air exchange rate, kitchen volume, stove burn time and so on. Through comparisons based on simultaneous measurement of emissions and pollutants in the South East Asian region, the method has been demonstrated to have moderate quality for linking ERs and IAQGs. Based on this approach, ER guidelines for $PM_{2.5}$ and CO result in an intermediate target of 60% and final target of 90% of households meeting the IAQGs. The annual final IAQGs of $PM_{2.5}$ (10 μg/m^3) and 24-h CO (7 mg/m^3) are used as chronic exposure levels for human health. WHO also provides ER targets for stoves with chimneys used for ventilation, assuming 25% of the pollutant emission from stoves escapes into the indoor environment. Therefore, the final and intermediate ER targets for vented stoves are calculated based on the IAQGs and MCBM (final ERs targets for CO and $PM_{2.5}$: 0.59 g/min and 0.80 mg/min; intermediate ER targets for CO and $PM_{2.5}$: 1.45 g/min and 7.15 mg/min).

Sampling and laboratory analysis

The emission exhaust was sampled using a hood 0.5 m above the cooking surface (stoves). A flue pipe was connected after the hood to vent smoke out of the laboratory (Fig. A.1). The sampling hood design and procedure are the same as that in a previous study (Carter et al., 2014). The sampling probes for gas temperature (Temperature Meter, DT-625, CEM, Shenzhen, China), PM and CO/CO_2 were placed into a small hole in the middle of the flue pipe. Real-time CO, CO_2, and CH_4 concentrations were measured online (GXH-3051, Junfang, Beijing, China). The gas monitors were calibrated for zero (pure nitrogen) and span (standard gases: 1.00%, 10.0%, and 0.1% for CO, CO_2, and CH_4) checked previously in the laboratory. Two active samplers (AirChek XR 5000, SKC, Eighty Four, PA, USA) were used to collect PM (including EC and OC) using quartz fiber filters (QFFs, 37 mm in diameters), and particulate PAHs using glass fiber filters (GFFs, 37 mm in diameters) followed by polyurethane foam plugs (PUFs, 22 mm diameter × 7.6 cm) to collect gaseous organics, such as PAHs. Before every test period, clean filters and PUFs were used to collect the pollutants in the background air in the laboratory and were measured as blanks which were subtracted from the exhaust levels.

Laboratory analysis of PM, EC, OC, and PAHs follows the procedure in previous studies (Shen et al., 2012a,b). Briefly, PM collected on filters was weighed by digital balance (0.01 mg) (Mettler Toledo XS105 DualRange, Columbus, OH, USA). EC and OC were measured by a Sunset EC/OC analyzer (Sunset Lab, Tigard, OR, USA). The procedure temperature was increased to 600 °C, 840 °C, and 550 °C in a pure helium atmosphere for OC detection, and then for EC detection at a temperature of 550 °C, 650 °C, and 870 °C in an oxygen/helium atmosphere. For particulate PAHs, including parent PAHs (pPAHs), nitrated PAHs (nPAHs), and oxygenated (oPAHs), the microwave accelerated reaction system (CEM, Mars Xpress, Matthews, NC, USA) was employed using 25 mL of *n*-hexane/acetone (1:1, v/v). The procedure temperature reached 110 °C within 10 min, and then was held for 10 min at 1200 W. For gaseous PAHs collected in PUFs, Soxhlet extraction was used at the temperature of 65 °C for 8 h, with 150 mL of the same mixture to microwave extraction.

All the extracts were concentrated to approximately 1 mL with a rotary evaporator (N-1100, EYELA, Tokyo, Japan) for purification. A silica/alumina column (10 mm diameter × 30 cm height) was used for purification. The column was packed with 12-cm height silica gel, 12-cm height alumina, and 1-cm height anhydrous sodium sulfate from the bottom up. Before elution, 20 mL of *n*-hexane was used to pre-elute the sample in the column. Then, a dichloromethane/n-hexane mixture (50 mL, 1:1, v/v) was used to elute the column. The elution was connected and concentrated into hexane solution, and spiked with 200 ng of internal standards, including naphthalene-d8, acenaphthene-d_{10}, anthracene-d_{10}, chrysene-d_{12}, and perylene-d_{12} for parent PAHs, and 1-nitroanthcene-d_9 and 1-nitropyrene-d_9 for derivative PAHs, all from J&K Chemical, Newark, DE, USA.

PAHs were analyzed in a gas chromatograph coupled with a mass spectrometer (GC-MS, Agilent 6890/5973, Santa Clara, CA, USA) and a DB-5MS capillary column (0.25 mm i.d. × 30 m, 0.25 μm film thickness). For parent PAHs, the electron ionization mode was adopted and helium was the carrier gas. The oven temperature was held at 50 °C for 1 min, then increased to 150 °C in 10 min, to 240 °C at a rate of 3 °C/min, and increased to 280 °C for 20 min. However, for derivative PAHs, a negative chemical ionization mode was adopted. High-purity helium and methane were used as the carrier and reagent gases, respectively. The oven temperature was programmed at 60 °C, and increased to 150 °C at a rate of 15 °C/min, and then to 300 °C at 5 °C/min, being held for 15 min. PAHs were identified and quantified based on the retention times and selected ions of standards shown above.

A total of 27 parent, 12 nitrated, and 4 oxygenated PAHs measured included acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLO), phenanthrene (PHE), anthrancene (ANT), fluoranthene (FLA), pyrene (PYR), benz(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz[a,h]anthracene (DahA), indeno(1,2,3-cd)pyrene (IcdP), benzo (g,h,i)perylene (BghiP), benzo[c]phenanthrene (BcP), retene (RET), perylene (PER), benzo(e)pyrene (BeP), coronene (COR), dibenzo[a,e] fluoranthene (DaeF), cyclopenta[c,d]pyrene (CPP), dibenzo[a,c]pyrene (DacP), dibenzo[a,i]pyrene (DaiP), dibenzo[a,l]pyrene (DalP), dibenzo [a,e]pyrene (DaeP), dibenzo[a,h]pyrene (DahP), 1-nitronaphthalene (1N-NAP), 2-nitronaphthalene (2N-NAP), 5-nitroacenaohthene (5N-ACE), 2-nitrofluorene (2N-FLO), 9-nitroanthracene (9N-ANT), 9-nitro-phenanthrene (9N-PHE), 3-nitro-phenanthrene (3N-PHE), 3-nitrofluoranthene (3N-FLA), 1-nitropyrene (1N-PYR), 7-nitrobenzo[a]anthracene (7N-BaA), 6-nitrochrysene (6N-CHR), 6-nitrobenzo[a]pyrene (6N-BaP), 9-fluorenone (9FLO), anthracene-9,10-dione (ATQ), benzanthrone (BZO), and benzo[a]anthracene-7,12-dione (BaAQ).

Quality control and data analysis

Procedure and reagent blanks were measured for every sample and subtracted from the results. The method detection limits (MDL) were 6.5–43, 5.8–121, and 21–57 pg/m^3 for gaseous pPAHs, nPAHs, and oPAHs, respectively; and 15–40, 5.0–89, and 3.3–66 pg/m^3 for particulate pPAHs, nPAHs, and oPAHs, respectively. 2-Fluoro-1,1′-biphenyl and p-terphenyl-d14 (J&K Chemical, Newark, DE, USA) were used as surrogate recoveries for pPAHs to monitor the quality of the analysis procedure. The surrogate recoveries (added randomly in 20% of the samples) for particulate and gaseous pPAHs were 82.7 ± 10.2%, 92.5 ± 13.0% and 76.7 ± 8.0%, 78.5 ± 13.7%. For derivative PAHs, 1-bromo-2-nitrobenzene (AccuStandard, New Haven, CT, USA) was used as the surrogate recovery. Those for particulate and gaseous derivative PAHs were 84.7 ± 17.0% and 81.7 ± 10.3%, respectively. The coefficients of variation for OTE, MCE, and HTE were 35%, 4.6%, and 24%, respectively.

Data statistical analysis was performed using the software SPSS 13.0 (IBM Corporation, Armonk, New York, USA), with the statistically significant level of 0.10. Kolmogorov–Smirnov *Z* (non-parameter) statistical test was used to compare levels between two series of samples. The correlation test was conducted through the non-parametric test of Spearman.

Results and discussion

Efficiencies and emission factors

Twenty-seven entire sampling cycles (from ignition to fire finishing) were conducted with three pellet-gasifier stoves, three testing phases (one phase with Chinese WBT, two phases with International WBT), and three replicates. As discussed in the Methods section, results were similar for cold- and hot-start phases of the International WBT, so average values are reported. Then, the efficiencies, flue gas temperatures (T), and EFs of various pollutants for different method–stove combinations are shown in Table 2 with arithmetic means and standard deviations. All the EF values are in units of mass of pollutant per mass of dry fuel. The measured EFs of target pollutants including CO, PM, OC, EC, 27pPAHs, 12nPAHs, and 4oPAHs (EF_{CO}, EF_{PM}, EF_{OC}, EF_{EC}, $EF_{27pPAHs}$, $EF_{12nPAHs}$, and EF_{4oPAHs}, respectively) are also listed in Table 3 in units of mass of pollutant per fuel energy. The values of EFs are also shown as arithmetic means and standard deviations for different sampling under various combinations in triplicate.

The values of OTE, MCE, and HTE (from individual test replicates) ranged from 16% to 44%, 79% to 99%, and 18% to 45%, respectively. Another study of pellet-gasifier stoves using the International WBT in the same testing laboratory reported that OTE was in the range of 17.9% to 33.3% (Carter et al., 2014), which was close to that in our present study, and comparable to those in a previous systematic study using the International WBT in U.S. EPA Cookstove Testing Laboratory (Jetter et al., 2012). The OTE of typical "improved" biomass stoves using unprocessed biomass in China, however, is usually less than 20% (Chen et al., 2010). As expected, the OTE for modern pellet-gasifier stoves are generally higher than those for improved biomass stoves, even though there are some exceptions under certain circumstances, and also much higher (p < 0.10) than those for traditional stoves, whose OTE are only about 10% when burning crop residue and firewood (Chen et al., 2010). Consequently, potential reductions in fuel consumption, and lower pollutant emissions for these modern stoves could be expected.

The EF_{CO} ranged from 0.182 to 12.3 g/MJ, with a mean and standard derivation of 2.40 ± 2.53 g/MJ. In units of mass of pollutant per dry fuel mass (g/kg), the average EF_{CO} for pellets measured in the present study was 41 ± 42 g/kg, ranging from 3.00 to 203 g/kg. By compiling available data in the literature, it was reported that EF_{CO} for crop residue pellet was about 21 g/kg (Shen and Xue, 2014). But, for the ordinary fuels like crop residues, wood logs, and wood branches burned in residential cookstoves, the EF_{CO} values were as high as 93, 53, and 120 g/kg, respectively, much higher (p < 0.10) than that for pelletized fuels.

The EF_{PM}, EF_{OC}, and EF_{EC} were in the range of 39.3 to 338 mg/MJ (0.673 to 5.78 g/kg), 1.05 to 8.08 mg/MJ (0.0174 to 0.124 g/kg), and 0.0264 to 2.99 mg/MJ (0.000452 to 0.0511 g/kg), respectively, and the average total carbon content of PM was about 2.4%. In a previous laboratory-simulated burning study, Boman et al. (2011) measured PM emissions for pellets at about 2–150 mg/MJ (per fuel energy). Shen et al. (2012c) reported that the EFs of PM from the burning of biomass pellets were 17.6–332 mg/MJ (per fuel energy) (Shen et al., 2012c), comparable to the results in this study. Jetter et al. (2012) reported that $PM_{2.5}$ EFs for biomass pellet cookstoves were 13–88 mg/MJ (per fuel energy) for cold-start and hot-start test phases of the International WBT, which were in the range of our study, though emissions of $PM_{2.5}$, not TSP, were reported. However, compared with the PM EFs for unprocessed/raw biomass fuels, the results were much lower (p < 0.10) for pelletized biomass fuels. Differences in both fuel properties and stove design are responsible for the significant difference found in emissions. The gasifier stoves have a supply of both primary and secondary air, which may improve efficiency in the modern burner compared to the uncompressed straw-burning in so-called improved brick stoves. The MCE calculated for pellet burning was generally higher than that of uncompressed straw burning.

For pPAHs, the $EF_{27pPAHs}$ were between 1.5 and 1900 μg/MJ, with a mean of 380 μg/MJ, of which the EFs of total 15 priority PAHs and BaP were 370 and 3.2 μg/MJ, respectively. For PAH derivatives, the EF_{4oPAHs} were 2.9–150 μg/MJ which was within the order of magnitude of pPAHs, while for nPAHs, the EFs were in the range of 0.046 to 4.3 μg/MJ, which was nearly 2–3 orders of magnitude lower than the pPAHs. In units of pollutant mass of per fuel mass (mg/kg), the overall average $EF_{27pPAHs}$, $EF_{12nPAHs}$, and EF_{4oPAHs} were 6.5, 0.024, and 0.77 mg/kg, respectively. Some previous studies, though limited, on PAHs emissions from the burning of pellets reported comparable results to those in this study. For example, the pPAH for pellets burned in a modern household stove ranged from 0.33 to 1.3 mg/MJ (Shen et al., 2012c), compared with pPAH EFs (0.0015 to 1.9 mg/MJ) in this study. The EFs of oPAHs were found to be in the range of 0.08 to 4.0 mg/kg (Shen et al., 2012d). Compared to uncompressed ordinary biomass fuels, the EFs of PAHs for pellets in this study were much lower (p < 0.10). The average pPAH and oPAH EFs for crop residues were reported to be about 63 and 8.1 mg/kg, respectively (Shen et al., 2011a,b). All the comparisons of the EFs and combustion efficiencies in this study and those of other previous studies were conducted under the process of statistical tests. However, large variations from various influencing factors still existed and large sampling size and reliable influencing factor control are needed in future studies.

In addition to total PAH EFs, the normalized composition profiles of individual compounds for pPAHs and their derivatives were also considered (Fig. A.2). For pPAHs, ACY, PHE, and PYR dominated the mass amount, comprising up to 50.6% of the total. The profile is very similar with those in previous studies on PAH emissions from pellet burning (Shen et al., 2012c; Boman et al., 2011). For the derivative compounds, 2N-NAP, 1N-NAP, 9N-ANT, and 3N-PHE were the predominating nPAHs, contributing over 83% of mass in total nitro-PAHs emission. The emissions of ketones (9FLO and BZA) were generally higher than that of quinines (ATQ and BaAQ). 9FLO was highest with the fraction of 43.8%, and followed BZA of 28.4%. The profiles of nPAHs and oPAHs are also similar to those from previous studies (Shen et al., 2012d, 2013).

The variances within the triplicate measurements are obviously lower than those between different method–stove combinations,

Table 2
Efficiencies (OTE, MCE, and HTE), flue gas temperature (T) and EFs of different pollutants for different method–stove (M–S) combinations are shown in units of pollutant mass per dry fuel mass. C and I represent the Chinese and International WBTs, respectively. Arithmetic means ± standard deviations are shown.

M–S	OTE	MCE	HTE	T, °C	CO, g/kg	PM, g/kg
C-stove 1	23 ± 3%	98 ± 2%	24 ± 3%	55 ± 5.7	20.2 ± 22.4	3.35 ± 0.63
C-stove 2	40 ± 2%	98 ± 1%	41 ± 2%	43 ± 3.1	17.5 ± 5.60	2.00 ± 1.18
C-stove 3	43 ± 1%	99 ± 1%	44 ± 1%	47 ± 2.0	14.6 ± 0.70	3.66 ± 0.58
I-stove 1	17 ± 1%	85 ± 7%	20 ± 1%	52 ± 0.1	145 ± 82.1	2.67 ± 0.09
I-stove 2	33 ± 1%	95 ± 2%	34 ± 1%	37 ± 2.1	47.4 ± 19.9	2.43 ± 0.94
I-stove 3	31 ± 5%	96 ± 2%	31 ± 4%	38 ± 2.7	40.6 ± 23.8	2.91 ± 1.45

M–S	OC, g/kg	EC, g/kg	∑ pPAHs, mg/kg	∑ nPAHs, mg/kg	∑ oPAHs, mg/kg
C-stove 1	0.023 ± 0.005	0.004 ± 0.002	6.54 ± 4.00	0.017 ± 0.005	0.47 ± 0.28
C-stove 2	0.031 ± 0.008	0.001 ± 0.001	0.11 ± 0.08	0.006 ± 0.007	0.10 ± 0.05
C-stove 3	0.028 ± 0.008	0.011 ± 0.005	0.76 ± 0.15	0.009 ± 0.005	0.64 ± 0.24
I-stove 1	0.043 ± 0.003	0.006 ± 0.002	1.70 ± 1.21	0.008 ± 0.002	0.21 ± 0.12
I-stove 2	0.077 ± 0.034	0.005 ± 0.003	1.51 ± 1.37	0.033 ± 0.015	0.44 ± 0.10
I-stove 3	0.087 ± 0.033	0.030 ± 0.016	16.7 ± 12.0	0.041 ± 0.021	1.69 ± 0.69

Table 3
EFs of different pollutants for different method–stove (M–S) combinations are shown in units of pollutant mass per fuel energy. C and I represent the Chinese and International WBTs, respectively. Arithmetic means ± standard deviations are shown.

M–S	CO, g/MJ	PM, mg/MJ	OC, mg/MJ	EC, mg/MJ	∑ pPAHs, μg/MJ	∑ nPAHs, μg/MJ	∑ oPAHs, μg/MJ
C-stove 1	1.22 ± 1.36	200 ± 40	1.41 ± 0.318	0.246 ± 0.107	397 ± 243	1.04 ± 0.300	28.5 ± 17.3
C-stove 2	1.02 ± 0.33	120 ± 70	1.80 ± 0.804	0.068 ± 0.598	6.00 ± 5.00	0.368 ± 0.408	5.95 ± 2.63
C-stove 3	0.86 ± 0.04	210 ± 30	1.64 ± 0.439	0.624 ± 0.302	45.0 ± 134	0.530 ± 0.284	37.3 ± 13.9
I-stove 1	8.77 ± 4.98	160 ± 10	2.63 ± 0.210	0.369 ± 0.104	103 ± 134	0.465 ± 0.101	12.9 ± 7.60
I-stove 2	2.77 ± 1.17	140 ± 50	4.49 ± 1.96	0.293 ± 0.151	88.0 ± 80.0	1.94 ± 0.860	25.8 ± 5.70
I-stove 3	2.37 ± 1.39	170 ± 80	5.07 ± 1.94	1.74 ± 0.914	977 ± 704	2.42 ± 1.23	98.6 ± 40.2

indicating great contribution to variances from different pellet-gasifier stoves and different test protocols. The difference between the two WBTs is detailed in the following sections.

Differences among three pellet-gasifier stoves

The differences in stove designs, such as control systems for primary and secondary air supply, are expected to result in different performance and subsequently distinct OTE, THE, MCE, flue gas temperature, and pollutant emissions. MCE can serve as a proxy for the efficiency of a stove. In comparisons of MCE among the three pellet-gasifier stoves, it was apparent that stove 1 had the lowest MCE compared to the other two stoves under both International and Chinese WBTs (Table 2). Fig. 3A compares the OTE and flue gas temperatures of these three stoves. As the temperature in the stove chamber was not measured in this study, flue gas temperature here is used to indicate relative difference in burning temperature among different test cycles, though potential uncertainty existed. Statistically higher ($p < 0.10$) flue gas temperature and lower OTE were found for stove 1 compared to the other two stoves under both International and Chinese WBTs. HTE which represents the ratio of useful energy (transferred to the water in the pot) to heat energy released from the fuel, and can be calculated from OTE and MCE (HTE = OTE/MCE), was compared. A significant positive linear correlation ($p < 0.10$) between OTE and HTE was found, as shown in Fig. 3B, and the lowest values are found for stove 1. Thus, OTE was a feasible parameter as HTE in this study, resulting from accurately controlled lab tests and relatively small differences in MCE.

Pollutant emissions for the three tested stoves are compared in Fig. 4. For CO, although the arithmetic means of EF_{CO} of stove 1 were higher than stoves 2 and 3, the difference was not statistically significant. The CO EFs for pellets burning in stove 2 and stove 3 were also similar. For PM, the lowest emissions were found for stove 2 ($p < 0.10$) in both International and Chinese testing protocols, and the ratio of EC to OC (EC/OC) was also the lowest in emissions from stove 2 ($p < 0.10$). The EF_{EC} and EC/OC of stove 3 were significantly higher ($p < 0.10$) than those of stove 1. Similarly, the lowest emissions of parent PAH emissions were also found in emissions from stove 2 ($p < 0.10$) under both protocols. The burning in stove 1 had much higher PAHs emissions than the burning in stove 3 by using the Chinese WBT protocol, but an opposite difference between stove 1 and stove 3 was observed when following the International WBT protocol. For PAH derivatives, a different comparison result was shown when using different testing protocols. By following the Chinese WBT protocol, the lowest emissions were found for stove 2, and the results were similar between stove 1 and stove 3. But when using the International WBT protocol, the highest emissions were found for stove 3, and those for the stove 1 were much lower. Different stove designs and testing protocols may result in a difference in conditions like combustion temperature and air–fuel mixing status during the combustion process, and thus result in different amounts of air pollutant emissions. Moreover, the influence on different chemicals varied due to distinct formation mechanisms. For better understanding, more studies are needed on detailed characterization of combustion processes and pollutant formation mechanisms.

Generally, the results showed that for most air pollutants, the lowest emissions were found for stove 2, regardless of which testing protocol was used. As mentioned above, stove 2 has an advanced inner structure design which can control the air mixing ratio and intensity of combustion ideally by controlling the ratio and gross flow rates of primary and secondary air supply under a precise and pre-set procedure, which is responsible for the low emissions found. For the comparison of stove 1 and stove 3, not only targeted air pollutants but also testing protocols should be considered if one hopes to select a suitable stove. A different testing protocol suggests how to operate the stove, which consequently affects the pollutant emissions, and the influence is different or even opposite for different air pollutants.

Evaluating indoor air quality through emission rate targets

Household solid fuel combustion can cause severe indoor air pollution. Linking the ER of pollutants of various stove–test combinations with indoor concentrations is a good way to evaluate the impact on

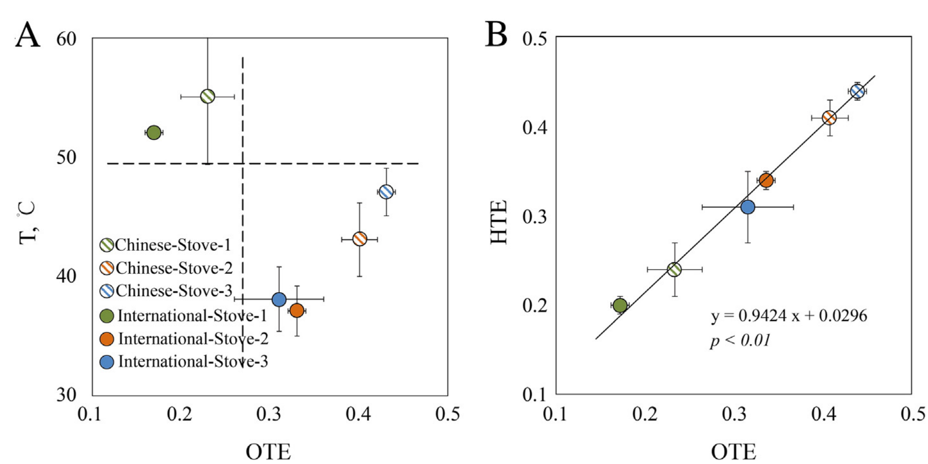

Fig. 3. The relationship between flue gas temperature (T) and OTE (**A**), and between HTE and OTE (**B**) were shown with *X*-, *Y*-axis standard errors.

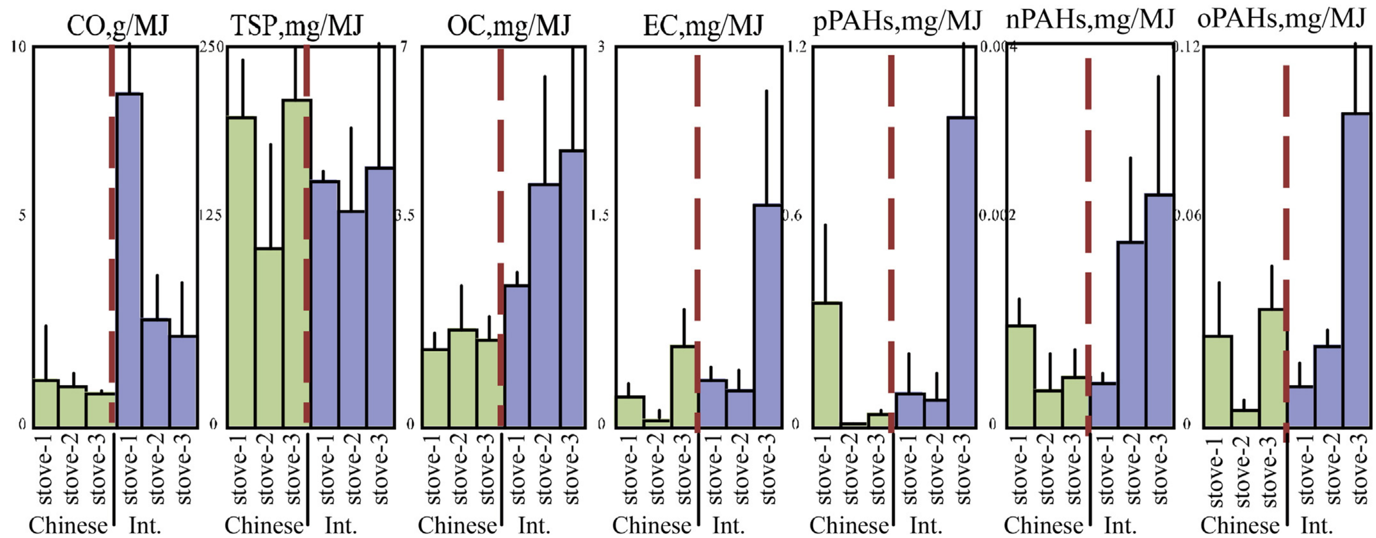

Fig. 4. Pollutant EFs of the three pellet-gasifier stoves with Chinese WBT and International (Int.) WBT are shown with standard deviations.

indoor air quality (IAQ). It can also provide guidance on what emissions performance levels are required for meeting the IAQGs. According to the recently released WHO IAQGs, the final and intermediate ER targets of CO and $PM_{2.5}$ are shown in the Methods section.

Based on measured pollutant emissions, recorded fuel consumption amounts and burning durations, we further calculated the ERs of CO and $PM_{2.5}$, and compared them to the ER targets in WHO IAQGs. Since $PM_{2.5}$ was not measured directly in this study, we estimated $PM_{2.5}$ ERs from PM ERs through the estimated mass ratio of $PM_{2.5}$ in PM. According to a previous study conducted by Bäfver et al. (2011) ($PM_{2.5}$ percentage in total PM was 84%–96%), the mass percentage was assumed at the level of 90% (Bäfver et al., 2011). CO and $PM_{2.5}$ ERs of various method–stove combinations are shown in Fig. 5, with the ranges of 18 to 210 mg/min and 0.12–7.1 g/min for $PM_{2.5}$ and CO, respectively. Both of them were higher than the final ER targets set by WHO IAQGs, and $PM_{2.5}$ ERs were much higher than the limit, compared to those of CO. A total of 29% and 58% of CO ERs exceeded the intermediate and final ER targets limits of WHO IAQGs, respectively, and all of $PM_{2.5}$ ERs exceeded both ER targets by approximate one order of magnitude. As

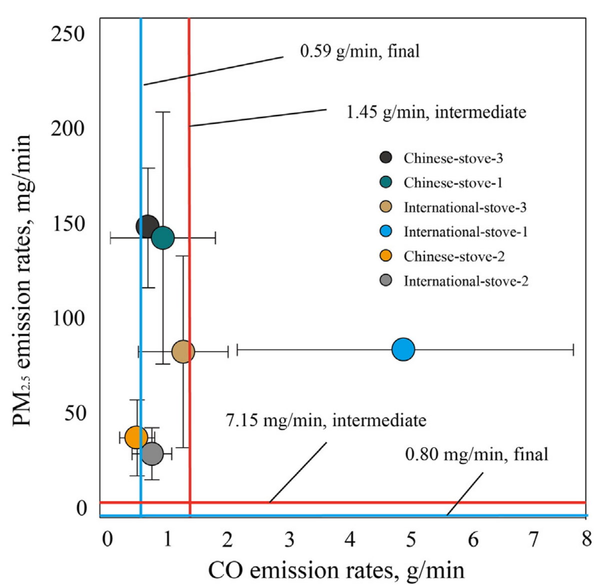

Fig. 5. CO and $PM_{2.5}$ ERs of six stove–test combinations were plotted with *x*-, *y*-axis standard error bars. Blue and red lines represent final and intermediate ER limits for $PM_{2.5}$ and CO, respectively.

indicated by WHO IAQGs, substantial improvement in $PM_{2.5}$ ERs is needed. A statistically insignificant correlation was found between CO and $PM_{2.5}$ in this study. It appeared that though the gasifier stoves did lower pollutant emissions compared to traditional stoves with unprocessed fuels previously existing in China, much more effort should be taken to reduce ERs to meet the IAQ standards. While it is recognized that the targets suggested by WHO IAQGs are also associated with uncertainties from model development, there is no doubt that the further improvement of stove technology would benefit air quality and human health.

Comparison between International and Chinese WBTs

In this study, emission experiments following the International and Chinese WBTs were conducted. It is interesting to compare the OTE, HTE, and pollutant EFs between these two different testing protocols. As shown in Table 2, OTE, MCE, HTE, and flue gas temperature (T) in the Chinese WBTs were significantly higher than those measured in the International WBT ($p < 0.10$). Compared with the Chinese WBT, the International WBT has a shorter testing period and higher average combustion powers in such a short time (Carter et al., 2014; Huangfu et al., 2013). With shorter test duration, there might be not enough time to heat the stove and chamber air, while in the Chinese WBTs, fuel burning usually lasts longer providing a longer test duration to heat the stoves, and subsequently leads to higher gas temperature and more efficient burning with significantly ($p < 0.10$) higher efficiencies (OTE, MCE, and HTE) (seen in Fig. 3 and Table 2).

The EFs of most air pollutants (except parent and derivative PAHs in stove 1) measured in the International WBTs were generally higher than those in the Chinese WBTs, and the observed difference was consistent among the three pellet-gasifier stoves. The differences for CO, OC, and EC for all the stoves were statistically significant ($p < 0.10$). As mentioned above, the average combustion temperature in the Chinese WBTs was higher than that in the International WBTs, which may lead to more efficient combustion, subsequently resulting in lower pollutant emissions. Another explanation for high pollutant EFs measured in the International WBTs compared to those in the Chinese WBTs is that the total duration for the burning cycle was longer in the Chinese WBT compared to that in the International WBT. It has been recognized that higher pollutant emissions occur during the lighting phase or initial phase (Roden et al., 2006; Carter et al., 2014; Chen et al., 2016) that was averaged into the calculation of EFs over the whole burning cycle. Therefore, in the Chinese WBT, a longer burning duration would lower the average EFs calculated over the burning period. It is of high interest to investigate the impact of pollutant emissions during the lighting phase on the overall average results in both laboratory and field tests. The relatively high emissions of parent and derivative PAHs in stove 1 cannot be well explained at this stage.

Higher emissions per useful energy were also found in tests following the International WBT, compared to the Chinese WBT. The results are as expected, since higher fuel energy-based emissions and lower thermal efficiencies were found in tests using the International WBT.

Similarly, ERs also varied between the two different testing protocols. The CO ERs for the International WBT were significantly higher ($p < 0.10$) than those for Chinese WBTs with the same gasifier stove due to the relatively shorter burning test duration of the International WBT. In addition, considering the combined influence of burning tests and stove types, stove 2 had the lowest emissions and highest efficiencies with both test protocols.

Implications and limitations

Residential solid-fuel incomplete combustion has been a major emission source of many types of air pollutants globally, especially in developing countries. Household air pollution has been identified as the top environmental health risk factor globally, and thus the development of clean fuels and clean stoves is of worldwide concern. However, there are still limited testing data on efficiencies and pollutant emissions from processed fuels and improved stoves. In this study, we measured and compared the OTE, HTE, MCE, and EFs of a variety of incomplete combustion products including CO, PM, EC, and PAHs, for three stoves using pellet fuel. The results showed that the stove with better control of primary and secondary air supply rates and mixing ratio had higher efficiencies and lower emissions of most incomplete combustion products. However, the CO and $PM_{2.5}$ ERs were still higher than the targets in WHO IAQGs.

It is widely accepted that in addition to fuel properties and stove type, many other factors like emission testing protocols affect stove performance. So far, there have been very few comparison studies between the International WBT—the most widely used protocol worldwide—and the Chinese WBT—a protocol commonly used in China. Statistically significant differences in efficiencies and pollutant emissions were found between the two testing protocols. The EFs of most air pollutants from the testing following the International WBT were higher than those using the Chinese WBT protocol, regardless of the stove type. The Chinese WBT may better represent the real cooking practice in China, but it may still need to be developed or updated by learning experiences from the International WBT which is developed based on abundant valuable studies of many researchers in this field and has been often used in many countries. A standardized international testing protocol should be developed as soon as possible. However, it must be accepted by multiple stockholders and more importantly, based on solid conclusive evidence from data in emission measurements. In addition, it is expected that pollutant emissions from solid fuel burning for space heating—another widespread activity in China besides cooking—could be evaluated following a well-designed test protocol in the near future.

As a well-controlled laboratory test with commonly accepted procedures, the WBT is widely used for evaluating performance of various fuel-stove combinations through boiling and simmering water. Though the WBT is repeatable to obtain reliable comparison results, some other field tests are also needed, such as the controlled cooking test (CCT) and kitchen performance test (KPT). Unlike the WBT, the CCT and KPT, can reflect the real fuel consumption and some characteristics of stove performance with local residents' operation (Dutt and Ravindranath, 1993; Controlled Cooking Test, CCT, 2004; Kitchen Performance Test, KPT, 2007; Bailis et al., 2007; Smith et al., 2007). Secondly, we only tested three pellet-gasifier stoves. With a relatively small sample size, though low pollutant emissions were observed, we need to conduct more tests on more stoves, especially those likely to be enrolled in future intervention programs. Furthermore, in the evaluation of potential impacts on health, we compared the ERs to the ER targets suggested by the WHO Guidelines. Pollutant emissions were only measured from the chimney exhaust, and the fugitive emissions are not included. A direct measurement of IAQ in real households may be more appropriate for the evaluation of potential health impacts in the use of these gasifier stoves.

Conclusion

To achieve better performance of stoves, advanced stoves with biomass pellet fuel have been promoted by some intervention programs. In this study, three popular commercial forced-draft pellet-gasifier stoves in rural China were tested using both the International and Chinese WBTs. Compared with traditional unprocessed biomass stoves in previous studies, not only the efficiencies but also the emissions of CO, PM, pPAHs, nPAHs, and oPAHs were improved. Better control of the ratio of primary and secondary air, as well as control of the gross air supply, under a precise and pre-set procedure appears to be critical for stove performance. However, the ERs of CO (18–210 mg/min) and $PM_{2.5}$ (0.12–7.1 g/min) did not meet the ER targets for $PM_{2.5}$ and CO suggested by the WHO IAQGs, being particularly too high for $PM_{2.5}$. This implies that much additional improvement of these pellet stoves will be needed if they are to meet WHO ER targets.

Efficiencies under the Chinese WBT were higher than those under the International WBT, and pollutant EFs using the Chinese WBT protocol were much lower. This could be explained by the relatively longer test duration of the Chinese WBT, which provides more time to heat the stove and leads to higher average gas temperatures, and also a long duration may lower the overall average emissions since high emissions are often observed during ignition. Different protocols represent somewhat different combustion/cooking activities and thus each could be said to be usable in parts of the world if the local practice is rather different. Their results cannot directly be compared. It may be necessary, however, to develop a hybrid WBT for international comparisons.

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Appendix A. Supplementary data

Some supplementary figures and tables are provided in the supporting material available free of charge via the Internet. Supplementary data associated with this article can be found in the online version, at doi:http://dx.doi.org/10.1016/j.esd.2016.02.008.

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