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Pollutant Emissions from Improved Coal- and Wood-Fuelled Cookstoves in Rural Households

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Supporting Information

ABSTRACT: Residential solid fuel combustion is a major source of many pollutants, resulting in significant impacts on air quality and human health. Improved stoves, especially some modern gasifier biomass models, are being deployed to alleviate household and ambient air pollution. Pollutant emissions from coal burning in improved metal stoves (n = 11) and wood combustion in modern gasifier stoves (n = 8) were measured in field in Hubei, China. The emissions of CO, TSP, OC, EC, and PAHs from coal burning in the improved iron stoves were generally lower than previously reported results for coal in traditional stoves. For pollutants from wood combustion in the gasifier stoves, the emissions were less than literature-reported values for wood burned in traditional stoves, comparable to those in improved stoves, but appeared to be higher than those for pellets in gasifier stoves in laboratory tests. The limitations of scarce data and large variances result in statistical insignificance. Daily emissions of targeted pollutants per household were found to be higher for wood burners, compared with households relying on coal. The gasifier stove had relatively high thermal efficiencies, but emissions of most air pollutants per delivered energy were not significantly different from those from the coal burning in improved iron stoves. Moreover, higher emissions of OC, EC, and PAHs were observed, indicating that caution and additional testing will be needed while designing future clean cookstove intervention programs.

INTRODUCTION

Resulting from large consumption amounts and relatively lower burning efficiencies, residential solid fuel combustion has been recognized globally, in particular developing countries, as a major source of many air pollutants, such as primary particulate matter (PM), black carbon (BC), and polycyclic aromatic hydrocarbons (PAHs). It has been estimated that globally, 21.4% of primary $PM_{2.5}$, 63% of PAHs, and 28.9% of BC are from the residential fuel combustion. In China, contributions of residential fuel burning to national total annual emissions are 36.1%, 62%, and 52.8%.[1−4] In the last several decades, although the proportion of solid fuel users has gradually decreased, this population remains very large and stable at about 2.8 billion, a fact that frequently raises public concerns.[5,6] The latest Environmental Exposure-Related Human Activity Survey in China found that about 43.4% of households used biomass and/or coal for daily cooking, and 29.5% of households relied on these solid fuels for heating in 2012, and these proportions were as high as 60.1% and 40.4% in rural areas.[7] Thus, residential solid fuel combustion leads to serious air pollution, not only of indoor but also outdoor ambient air.[6,8−10] Household air pollution has been widely reported as one of the top environmental risk factors, leading to no fewer than 4.3 million premature deaths in 2012 alone.[6,11] Besides obvious adverse health outcomes, pollutants emitted from the residential sector have notable impacts on local and regional climate.[12,13] One important compound is BC. BC is thought of as the second or third most important climate forcer, and recognized as the one that should be controlled to slow global

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warming in the short term.[14] It has been documented that biofuel burning from residential cooking contributes significantly to the brown clouds over South Asia.[15]

Efforts to reduce pollutants from residential fuel combustion have been made over the last several decades. These efforts have included the introduction of highly efficient or so-called improved stoves, along with cleaner energies like electricity, biomass pellets and gas resources (such as biogas, liquefied petroleum gas, and natural gas). In China, during the 1980s to mid 1990s, clean cookstove deployment was introduced in the national five-year plan, and the world-famous China National Improved Stove Program was initiated. Stoves deployed during this period generally had a thermal efficiency of around 20%, which was higher than that for a traditional stove of only 10% (and in many cases, less than 10% in reality).[16] Less fuel consumption is expected after the adoption of improved stoves. For instance, a field survey reported that the average daily use of crop straws decreased from 7.5 kgce (kg coal equivalent) to 2.5 kgce.[17]

From the late 1990s, the development of household stoves came into a new stage of technical innovation. One widely known is gasifier stoves, which usually have a secondary air supply at the top and primary air supplied from the bottom. These gasifier stoves are often classified as "high efficiency, low emission stoves" which are defined as those with thermal efficiency >35% for cooking, with emissions of smoke, SO_2, NO_X, and CO less than 50 mg/m^3, 30 mg/m^3, 150 mg/m^3, and <0.2%, respectively.[16] In our previous study, pollutant emissions from one gasifier burner burning straw pellets and wood pellets were measured and compared to the emissions from uncompressed fuel combustion in an improved brick stove.[18,19] Recently, Carter et al., reported thermal efficiencies and CO_2, CO, $PM_{2.5}$ emissions from wood pellet burning in four different gasifier stoves.[20] From the literature, it was found that the thermal efficiencies for gasifier stoves may range from 22% to 41% in laboratory Water Boiling Tests, depending on fuel-stove types and burning conditions.[16,20−22] These data are very important and valuable in the evaluation of gasifier stove performance. However, such studies are still very scarce, with limited data on not only stove thermal efficiency, but also pollutant emissions. Moreover, to our knowledge, there is no field based measurements on pollutant emissions from gasifier stoves in China. Field studies are generally accepted to be closer to actual emission status of stoves in use, providing reliable data for the design and evaluation of potential intervention programs.

In the present study, field emission tests were conducted in rural Hubei Province, where biomass gasifier stoves have been introduced in a portion of rural households, while other residents still use improved coal stoves. The objectives of this study are 1) to obtain emissions of pollutants including CO, TSP, OC, EC, and PAHs, from the modern gasifier stove burning wood fuels based on a field test, 2) to compare pollutant emissions for the gasifier stove with those in improved and traditional stoves, and 3) to compare emissions between coal combustion in improved stoves and wood burning in gasifier stoves to gain insight into future intervention programs for changing from coal use to biofuels.

MATERIALS AND METHODS

Site and Fuel-Stove Information. The field measurement was conducted in rural Enshi County of Hubei Province (Supporting Information Figure S1). This county is in a mountainous zone, with about 75% of the population living in rural areas. With abundant coal resources (exploitable yield about 1.2 million tonnes/year),[23] coal consumption make up to 68% of total annual energy consumption in the region. Given strong supports from the local government and clean fuel/stove intervention programs, many residents are adopting modern gasifier stoves burning woody materials for daily life. It is reported that in 2013, over 20 500 households adopted high-efficiency biomass stoves.[24]

In the studied village, the coal stove and biofuel gasifier stove were deployed to rural households under the support of local village committee in 2008 and 2010. These two stoves are similar in appearance (about 70 cm in height with the diameter of inner burning chamber at about 30−40 cm and equipped with an outdoor chimney, Supporting Information Figure S2), but directed by the manufacturer to be used for the burning of coal and biofuels, respectively, with different designs of stove inner chamber. Fuels are top-loaded. A secondary side air is supplied for the gasifier stove to promote the burning of biofuels, and the fire power was about 3.0 kW while cooking.

Coals and woody fuels used in households were collected and taken back to the laboratory for elemental analysis and proximate analysis (Supporting Information Table S1). The volatile matter contents of coal and wood fuels were 8.67% and 85.6%, and fuel moistures were 1.35% for coal and 5.75% for wood fuels. The coals used in the studied region were anthracite ones, but it is important to note that nowadays in rural China, bituminous coals are still frequently used in many other regions.

Field-Based Kitchen Performance Test (KPT). Daily fuel consumptions in households were quantified by conducting the KPTs over an extended period of 3 days.[25] In households who were selected randomly in the county and willing to participate into the fuel consumption measurement study, enough fuel was prepared and weighed using an electric balance with the precision of 20g on the first day of the test (Supporting Information Figure S3). The families were asked to only use the preweighed fuels during the study period. Three days later, each household was revisited one-by-one and the remaining fuels were weighed. A total of 84 households using coals and 84 households using wood participated in the field KPT study.

Sampling and Analysis. Emission tests were conducted during the lunch hour in households who were willing to open the kitchen to us and would cooperate in emission tests. Residents were asked to conduct fuel burning and cooking activities as normal. Such field tests are often time-consuming, high labor cost, and particularly need high collaboration of household members. Finally, a total of 19 households (11 using coals and the other 8 burning wood) participated in the present field emission measurement study. The stove tested was the primary one used for daily cooking. No household reported a repair of stove during their usage. The field emission measurements were done by four groups and finished in 1 week. So, it might be expected that the weather and wood humidity conditions were similar for the different test cycles.

In the emission tests, the sampling probes were placed near the center of chimney exit. No further dilution was performed. Quartz fiber filters were used to collect total suspended particles (TSP), and were analyzed for OC and EC contents.[26] Glass fiber filters followed by polyurethane foam plugs (PUFs; 22 mm diameter × 7.6 cm, density 0.024 g/cm^3) were used to collect particulate and gaseous-phase organic compounds. Flow rate was ∼1.5 L/min, and was calibrated before and after each

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Table 1. Average, Standard Deviation (stdev) and Geometric Means (GM) of Emission Factors of Targeted Air Pollutants (Per Fuel Mass, As Received) From Residential Coal and Firewood Burning and Statistical Results of Data Comparison between Coal and Wood

	coal chunk (N = 11)			wood (N = 8)			
fuel type	average	Stdev	GM	average	Stdev	GM	p value[b]
CO_2, g/kg	1271	84	1269	1445	57	1444	0.696
CO, g/kg	58.1	45.2	37.1	90.2	30.7	84.3	0.294
OC, g/kg	0.145	0.177	0.090	1.72	1.38	1.14	0.003*
EC, g/kg	0.017	0.023	0.0059	0.280	0.143	0.244	0.001*
TSP, g/kg	2.45	2.62	1.52	3.16	1.50	2.84	0.127
22 pPAHs[a], mg/kg	26.0	30.4	11.0	74.2	50.1	57.4	0.108
BaP, mg/kg	0.053	0.072	0.017	0.308	0.304	0.200	0.056
9 nPAHs[a], mg/kg	0.190	0.148	0.146	0.236	0.073	0.226	0.492
4 oPAHs[a], mg/kg	0.455	0.475	0.293	4.14	1.44	3.90	0.0003*

[a]EFs of the parent and derivative PAH individuals are listed in Supporting Information Table S2. [b]Statistical test results (2-tails) of difference between coal and wood combustion emissions in term of per fuel mass based pollutant emissions by using the nonparameter Kolmogorov−Smirnov test with a significance level of 0.05.

sampling cycle using a primary flow calibrator (Bios Defender 510). Before and after each sampling cycle, PUFs and filters were packed and sealed separately in clean aluminum foil bags. The CO and CO_2 in emitted exhaust were measured using a nondispersive infrared sensor (GXH-3051, China). The instrument was calibrated using a span gas in the laboratory prior to use in the field and zero-checked before each field sampling cycle.

The TSP was gravimetrically weighed using a high precision digital balance (0.01 mg, XS105, Mettler-Toledo). Before weighing, the filters were equilibrated in a desiccator for 24 h in the room with ambient temperature of 25−30 °C and relatively humidity of 45−55%. Carbonaceous fractions (OC and EC) in TSP were analyzed using an OC/EC analyzer (Sunset Laboratory, Tigard, OR), with temperature protocols of 600, 840, and 550 °C in pure helium for OC detection, and 550, 650, and 870 °C in an oxygen/helium atmosphere to quantify EC. Pyrolyzed OC was estimated according to transmitted light intensity and subtracted from the EC result.

To analyze parent and derivative PAHs, the PUFs were exacted by 150 mL dichloromethane for 8 h, and particle-loaded filters were extracted with 25 mL hexane/acetone mixture using a microwave accelerated system (CEM Mars Xpress, Matthews, NC). The MAE system was operated at 1200 W (100%), and the temperature protocol was: increased to 100 °C in 10 min and held for another 10 min. After extraction, the solvent was concentrated to ∼1 mL and then transferred to a silica/alumina gel column for cleanup. The column was pre-eluted with 20 mL hexane (discharged) and then a 70 mL hexane/dichloromethane mixture. The eluate was concentrated to 1 mL and spiked with deuterated internal standards (J&W Scientific).

PAHs were analyzed by a gas chromatograph/mass spectrometer (Agilent, Santa Clara, CA) with an HP-5MS capillary column using an optimized temperature program and in electron ionization mode for parent PAHs and chemical ionization mode for derivatives.[27] Parent and derivative PAH individuals were identified based on retention time and qualifying ions of standards in selected ion monitoring mode, and quantified using selected quantitative ions (m/z) for each individual. A total of 22 parent PAHs, nine nitrated PAHs and four oxygenated PAHs (Supporting Information Table S2), denoted as 22pPAHs, 9nPAHs, and 4oPAHs, respectively, were addressed in the study. Detailed information about the analysis condition is present in the Supporting Information.

All filters were baked at 450 °C for 6 h and equilibrated in a desiccator. Before sampling, PUFs were pre-extracted with acetone, dichloromethane, and hexane in sequence (8 h each). Field and laboratory blanks were measured and subtracted from the samples. Instrument detection limits, method detection limits and recoveries from spiked standards were measured in preliminary experiments[26] and provided in the Supporting Information.

Data Analysis. Emission factors (EFs) of CO_2, CO, TSP, OC, EC, pPAHs, nPAHs, and oPAHs (EF_{CO2}, EF_{CO}, EF_{TSP}, EF_{OC}, EF_{EC}, EF_{pPAHs}, EF_{nPAHs}, and EF_{oPAHs}, respectively) were calculated based on the carbon mass balance method. The method has been widely used in emission measurement studies and gives comparable results to those from the whole collection in a lab chamber.[28−30] Modified combustion efficiency (MCE), defined as $CO_2/(CO+CO_2)$ (molar basis) was calculated to quantitatively describe combustion efficiency. Statistical analysis was done using STATISTICA software with a significance level of 0.05. The nonparameter *Spearman* and *Kolmogorov−Smirnov* tests were adopted.

A ratio (R), defined as $1\text{-}EF_{E\text{-}wood}/EF_{E\text{-}coal}$, was calculated to directly compare pollutant emissions between the coal and wood combustion. A positive value may indicate possible reductions in pollutant emission when coal use in improved stoves was replaced by wood in gasifier stoves. The Monte Carlo simulation running 10 000 times was conducted to address uncertainties in the estimation. With obtained information on the distribution of estimated R values, the results are expressed as a median and an interquartile range.

RESULTS

Measured EFs of targeted pollutants for coal and firewood are summarized in Table 1. Results for individual parent and derivative PAHs are provided in detail in Supporting Information Table S2. The EFs are reported in the unit of pollutant mass per fuel mass on a wet basis. EFs on a dry basis are also calculated and listed in Supporting Information Table S3.

In particles from the coal combustion, the carbonaceous fractions (EC+OC) comprised only 9.8% of total particle mass. The mass ratio of EC to OC was only 0.094. Among the 22 parent PAHs measured, naphthanene, phenanthrene, acenaph-

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thylene, and pyrene were major components. EFs of individual nPAHs were similar, at about 10−30 μg/kg. For individual oPAHs, 9-fluorenone was the most abundant one with an EF of 226 μg/kg, and benzo[a]anthracene-7,12-dione was only 20 μg/kg. The gaseous pPAHs, nPAHs and oPAHs contributed to 89%, 73%, and 90% of total pPAHs, nPAHs, and oPAHs emitted from the coal combustion, respectively.

For the firewood burning, total carbon accounted for as much as 54% of the particle mass, which was much higher than that from the coal combustion. The EC/OC ratio was as high as 0.34. Approximately 87%, 64%, and 80% of parent, nitrated and oxygenated PAHs were in the gaseous phase. The EF of retene, which is a specific marker for softwood, was 1.27 ± 0.97 mg/kg. Again, naphthanene, phenanthrene, acenaphthylene, pyrene, and fluoranthene were dominant parent PAHs. Individual nPAHs were comparable to each another, in the range 12−46 μg/kg. 9-fluorenone was the most abundant oPAH identified with an EF of 2.67 mg/kg, and that of benzo[a]anthracene-7,12-dione was 85 μg/kg.

It is notable that in both coal and firewood burning emissions, measured air pollutant EFs strongly varied. Coefficients of variation (COVs) in the EFs for the coal were 80−140%, and 30−80% for the firewood combustion. In a field measurement on residential wood combustion in Honduras, calculated COVs in the EF_{TSP}, EF_{OC}, and EF_{EC} were 13%−28%, 18%−30%, and 11%−33% for a specific fuel/stove combination, and the overall average COVs were 19%, 23%, and 20%, respectively.[31] A previous field study on indoor crop straw burning reported that the COVs in EF_{TSP}, EF_{OC}, and EF_{EC} were 34−81%, 29−145%, and 30−117% within the triplicate measurements of a fuel/stove combination, and at 63%, 112%, and 109% on average.[32] In another field study, the COVs in EF_{TSP}, EF_{OC}, and EF_{EC} were 42%, 52%, and 76% for the indoor crop straw burning, and 27%, 35%, and 46% in the residential wood combustion.[33] The variances in pollutant emissions are attributable to many factors including fuel properties, stove types, fire management behaviors and also uncertainties in chemical analysis.[34,35] The latter two are often key factors affecting EFs for a specific fuel/stove combination. These key influencing factors should be well investigated, not only in field, but also in laboratory study, to get comparable results, and taken into account when addressing the uncertainties in emission inventories.

DISCUSSION

Correlations among Targeted Pollutants. The MCE values for the coal and wood combustion were 93.1 ± 5.4% and 89.8 ± 4.0%. As expected, positive correlations were found between MCE and CO_2 EF, and negative correlations between MCE and CO EF ($p < 0.05$). The relationship between MCE and other measured pollutants was statistically insignificant, though showing negative tendencies.

For both coal and firewood burning, there was no significant correlation between CO and other incomplete burning pollutants. CO is sometimes regarded as an indicator, or surrogate, of incomplete combustion process. However, either positively or negatively correlated or uncorrelated relationships have been mentioned in literature. The formation mechanisms and pathways usually vary among different compounds. Since these pollutants have different environmental and health impacts, the use of CO as a single index indicating fuel burning and pollutant emission status might be inadequate.

PAH individuals were positively correlated with one another in both coal and firewood combustion emissions, with an exception of retene. Retene is a specific PAH that is often considered a tracer for softwood burning.[36] In this study, retene was also detected in coal emissions, though a much higher level was found in emissions from wood combustion. In coal combustion, retene was positively correlated with PAHs like fluoranthene, pyrene and phenanthrene, but in emissions from the wood burning, there was no correlations found between retene and other PAHs. This information would be helpful for the evaluation of retene as an indicator for softwood burning.[37]

Considering the relationship between PAH derivatives and corresponding parent PAHs, statistically positive relationships were found between those in emissions from coal combustion, but insignificant relationships for those from the firewood burning. This might be related to differences in fuel properties (i.e., carbon and hydrogen contents) and environmental conditions (i.e., combustion temperature, relative humidity, and levels of free radicals) which are both responsible for the formation and changes of these trace organics in solid fuel burning. Unfortunately, these were not monitored in the present study. It would be interesting to examine formation mechanisms and potential influences of these organic compounds through online measurements using advanced instruments, such as high resolution mass and infrared spectrometers.

PAHs and their derivatives are emitted in gaseous and/or particulate phases. Usually, low molecular weight PAHs are more volatile and largely found in gaseous phase, whereas high molecular weight ones tend to be present in particulate phase. Compared with parent PAHs, volatilities of derivatives are lower because of attached polar functions, and therefore have a stronger tendency to be associated with particles. The partitioning of organic compounds is expected to be controlled by absorption, adsorption, or both, and affected by factors like the presence of absorbents and adsorbents, mixing status, temperature, relatively humidity and others. The partitioning can be described using the partitioning coefficient (Kp), calculated as $F/PM/A$, where A and F are gaseous and particulate phase organic concentrations, and PM is particle mass concentration.[38] In both the coal and firewood combustion emissions, the calculated Kp values are found to be positively correlated with the octanol-air partition coefficient (K_{OA}) and negatively correlated with subcooled liquid−vapor pressure (P_L^0)(with slopes −0.10 and −0.40, respectively), indicating that the partitioning of PAHs in emission exhaust is mainly governed by organic absorption rather than by adsorption.[39,40] The absorption-dominated partitioning of organic compounds in fresh emissions have also been found previously.[27,41] It is accepted, however, that once emitted into the air, the partitioning of these compounds changes during the aging process. The mass percentages of parent PAHs were only 0.1−0.5% of the TSP mass, and the PAH derivatives contributed a very smaller mass fraction of the particle. However, it is important to highlight the adverse health impacts of these pollutants, especially the derivatives which have been considered to have greater toxic potentials.

Comparison with Results in the Literature. In this section, we compared our results to data documented in literature with main focus of the performance of different fuel/stove types. Pollutant emissions vary dramatically among different fuel-stove combinations, affected by fuel properties and stove design.[35,42,43] The differences in experiment design

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Table 2. Average EFs of CO, TSP, OC, EC (g/kg) and Beozo[a]pyrene (BaP, mg/kg) from Residential Solid Fuel Use in Traditional and Improved Stoves in China[a]

fuel	stove	EFs	CO	TSP	OC	EC	BaP
anthracite chunk	traditional	mean	104	2.46	0.184	0.0123	0.380
		Stdev	42.4	3.05	0.248	0.0138	0.296
		GM	98.6	1.25	0.0896	0.0073	0.317
		N	4	7	3	3	2
wood	improved	mean	53.6	2.24	0.791	0.634	0.142
		Stdev	21.2	1.39	0.804	0.646	0.140
		GM	50.8	1.91	0.522	0.408	0.095
		N	25	28	27	27	24
	traditional	mean	89.1	6.06	2.91	1.25	4.87
		Stdev	56.7	3.92	1.01	0.479	1.51
		GM	70.5	4.74	2.82	1.20	4.75
		N	3	3	2	2	2

[a]Data shown are calculated arithmetic means (mean), standard deviation (stdev), and geometric means (GM) from collected data (sample size-N is also listed) in literature.

(i.e., Water Boiling Test or Controlled Cooking Test), fire management behaviors, sampling and laboratory methodologies also contribute to the variances, in addition to the fuel and stove types.

As mentioned above, emission tests on gasifier stoves are very scarce in China and two available studies were conducted in a laboratory and burned biomass pellets.[18,20] The emissions of $PM_{2.5}$ measured by Carter et al., ranged from 0.124 ± 0.027 g/MJ to 0.549 ± 0.135 g/MJ, varying among different stove types and ignition conditions.[20] The EF_{TSP} values for wood pellets and corn straw pellet in a gaisifer burner with secondary air supply in our previous study were 0.112 ± 0.048 and 0.204 ± 0.084 g/MJ.[18] These results appear to be lower than the measured emission factors in the present study. Among various influencing factors, the differences in fuel forms (wood pellet vs raw wood fuels) and stove types are thought to be main contributors.The present filed study measuring pollutant emission from wood burning in gasifier stoves merits more future studies to fill the research gap.

We compiled available data in literature on pollution emission factors from residential solid fuel combustion in China, and classified these data into different fuel and stove types (traditional vs improved), as shown in Supporting Information Table S4. Beside wood and anthracite chunk fuels tested in the present study, data on other fuel types including crop straw, branches, bituminous coals, and coal briquettes are included as well. The results for wood and anthracite chunk are also provided in Table 2, so as to have a clear comparison with results from the present study (Table 1).

For coal, most available studies reported emissions from bituminous coals, with very small numbers of data available for anthracite. This could be due to the fact that in a national scale, much more bituminous coals are consumed in the residential sector. For anthracite coals, the data available in literature was from the burning in traditional stoves. The average EF_{TSP}, EF_{OC}, and EF_{EC} values for anthracite chunk in traditional stoves are close to the results for anthracite burning in improved stoves in this study, but the emissions of CO and BaP for wood combustion in improved stoves were obviously lower.However, the differences were statistically insignificant, which might be due to limited sample sizes and large variances in emission measurements.

For wood combustion, the emissions in the gasifier stove in present study appear to be lower than those for wood burning in traditional stoves, and comparable to the results for wood combustion in improved stoves. The statistical analysis revealed insignificant differences ($p > 0.05$), which again might be attributable to large variances in emissions and much small sample sizes in each group. For example, there were only three data sets available for the emissions from wood burning in traditional stoves. In addition to the limitation of emission variances and sample size, the insignificant difference between emissions from the gasifier stove in this study and the results in improved stoves might be also attributable to the speculation that the wood combustion in gasifier stoves in real use were not as efficient as expected, even though previous laboratory tests had reported relatively higher thermal efficiencies and lower pollutant emissions in WBT test by burning pellets in gasifier stoves. Without enough data support, it is impossible to reach a sound conclusion at this stage, and more studies in the future are expected.

COAL-VERSUS WOOD-FUELLED COOKSTOVE EMISSIONS

In the studied area, coal burning in improved stoves is very common, but wood combustion in gasifier stoves is proposed because of its higher thermal efficiency. To provide direct information on pollution control and health protection strategies for policy makers, it is of high interest to compare pollutant emissions for these two fuel-stove combinations. To do such comparison, it is more appropriate to use delivered-energy based pollutant emission factors (EF_E) calculated as EF/η/HV, where η and HV are burning thermal efficiency and fuel net calorific value, respectively. Laboratory tests at the stove testing center showed that thermal efficiencies for the coal-fuelled improved stove and biofuel gasifier stove were 20% and 37%, respectively. In the present study, stove thermal efficiency was not measured in the field. So, the results in previous lab tests are adopted in herein calculation, which will led to considerable bias in estimating energy-based emissions. For instance, if thermal efficiency of the biomass gasifier stove was not as high as 37%, the calculated energy-based emissions would be underestimated.

Figure 1 compares per delivered energy-based emissions of the targets between the coal and wood combustions. The CO

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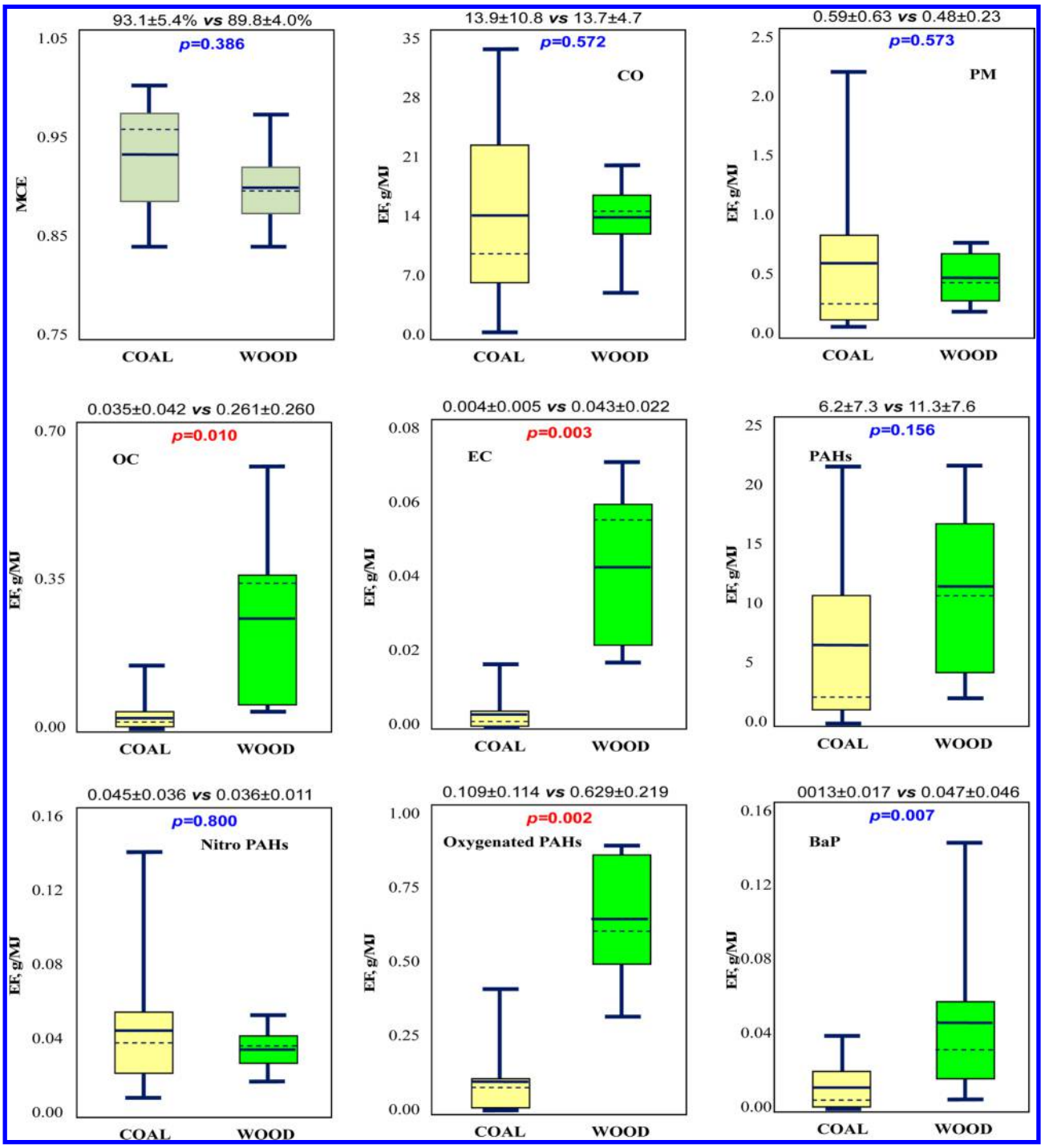

Figure 1. Comparison of MCE and per delivered energy based emissions of incomplete air pollutants (CO, PM, OC, EC, parent PAHs, BaP, nitrated PAHs and oxygenated PAHs) between coal and wood combustions. Data shown in the box plot are the minimum, maximum, the 1st and 3rd quartile, median (broken line) and arithmetic average values.

Table 3. Ratio (*R* Values) Of Delivered Energy Based Emission Factors (EF_E) of Pollutants from Wood Combustion in Comparison to Those in Coal Combustion

	CO	OC	EC	TSP	28pPAHs	9nPAHs	4oPAHs
average	−2.38	−16.2	−89.7	−0.971	−10.2	−0.365	−12.9
median	−0.20	−7.45	−19.5	−0.354	−2.27	0.110	−7.11
the 1st interquartile	−1.54	−21.2	−80.4	−1.86	−10.1	−0.790	−16.4
the 3rd interquartile	0.407	−1.40	−4.94	0.527	0.0159	0.385	−3.15

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Table 4. Estimated Daily Emissions of CO, OC, EC, TSP (g/day/capita) and PAHs (mg/day/capita) in Households Using Coal in Improved Stoves and Wood in the Gasifier Stove[a]

		CO	OC	EC	TSP	28pPAHs	9nPAHs	4oPAHs
coal user	mean	228	0.579	0.066	9.82	104	0.753	1.81
	median	209	0.524	0.060	8.99	94.8	0.684	1.66
	R_{50}	94.0−345	0.091−0.123	0.004−0.123	2.30−16.5	19.1−180	0.305−1.14	0.467−3.00
wood user	mean	444	8.55	1.39	15.4	367	1.17	20.5
	median	428	8.14	1.32	14.8	347	1.13	19.8
	R_{50}	313−560	3.63−13.0	0.841−1.87	9.65−20.5	183−529	0.847−1.45	14.4−25.7

[a]The results are calculated from measured EFs and obtained fuel consumption amounts through field fuel survey. Data shown are means, median values, the 1st and 3rd inter-quartile range (R50).

and PM appeared higher in the coal combustion, but statistically insignificant. Much lower emissions of OC and EC were found in the coal combustion relative to the wood burning (p = 0.010 and 0.002, respectively), and the EC/OC ratio was larger for the wood than that for the coal. Higher carbon fractions in particles from the wood combustion are related to higher carbon content in biomass fuels compared with fossil fuel, and relatively higher burning temperatures in the gasifier stoves which may be favorable for the formation of light absorbing particles. For the parent PAHs and derivatives, higher emissions of the 22pPAHs, BaP, and 4oPAHs (11.3, 0.047, and 0.629 mg/MJ, respectively) were found for the wood, compared with 6.2, 0.013, and 0.109 mg/MJ for the coal (p=0.156, 0.007, and 0.002, respectively). Table 3 lists the calculated ratios (R) of pollutant emissions between the coal and wood combustions. A wide range of estimated R value could be found, which is mainly due to relatively large variances in measured EFs. Generally, the ratio values are negative, indicating probably high pollutant emissions from the wood combustion relative to the coal burning.

The replacement of coal by wood is expected to yield obvious benefits regarding climate change. Relatively higher emissions of light-absorbing carbon in particles from wood combustion, however, may interfere with efforts toward CO_2 emission reduction to control warming, since there is increasing evidence that EC can enhance warming of the environment. The calculation of Global Warming Potential (GWP) Metrics is a helpful and interesting way to quantitatively assess potential impacts on climate of these pollutants. Following the method previously used by Zhi et al.,[44] we estimated the GWPs of light absorbing BC in the present paper as an example. Since we did not measure BC in this study, the results of EC were adopted in the calculation and comparison of GWP. An average $GWP_{BC,20}$ of 2200 was used (the subscript 20 means for a time scale of 20 years).[45] As a result, an increase of about 38.5 mg BC per useful energy delivered (from an average of 3.99 mg/MJ in the coal combustion to 42.5 mg/MJ in the wood combustion) would be equivalent to 84.7 g CO_2, which was obviously lower than 304 g/MJ CO_2 measured in emissions from the coal combustion. It is necessary to note this is a very simple and rough estimation with uncertainties attributable to variances in pollutant emission factors and the GWP values. A future study with more abundant data and more targets (like BC, OC, and sulfate) included into the estimation is necessary to reach a sound conclusion on the potential change in their environmental impacts through fuel and stove intervention programs.

Since the stove thermal efficiency was not measured in the field, a comparison between the wood and coal combustions might be associated high bias in potential. Another approach to approximate this data is to estimate emissions directly based on the measured EFs and fuel consumption amounts from the KPT study. The results from a preliminary KPT study indicated that daily fuel consumption per capita was about 3.96 ± 1.18 kg in households using coal, and 4.93 ± 1.13 kg in households relying on wood. By adopting conversion factors of 0.571 kgce/kg for wood and 0.7143 kgce/day for raw coal,[46] the daily fuel consumptions in households burning coals were 2.83 ± 0.84 kgce, and were 2.82 ± 0.65 kgce for households using wood. Accordingly, the daily emissions are estimated and listed in Table 4. Much higher emissions of targeted air pollutants were observed in households using wood in gasifier stoves compared to those households burning coals.

With relatively higher thermal efficiencies for gasifier biofuel stoves, lower fuel consumption amounts are often expected per cooking task. From the field survey, however, it was found that the daily fuel consumption in households using wood did not decrease significantly. In addition to relatively lower heating value of wood fuels, another important reason might be that the actual thermal efficiency may be not as high as expected, as mentioned above. Also, thermal efficiency may decline with aging from stove use, and available data on stove thermal efficiencies are from laboratory tests of new stoves. Unfortunately, we did not measure actual thermal efficiency. In addition, it has been recognized that an increase in stove thermal efficiency does not necessarily mean an enhancement of fuel combustion efficiency. Relatively high pollutant emissions have been reported for some improved stoves with higher thermal efficiencies that were deployed during the 1980s in China.[47] In fact, the MCE in the present wood combustion was 89.8 ± 4.0%, apparently lower than that of 93.1 ± 5.4% in coal burning (but statistically insignificant, p = 0.386). In certain circumstances, stoves or burners are designed to reduce heat loss and to increase thermal efficiency, with less attention paid to the impact on fuel burning. This highlights the future importance of simultaneously measuring stove thermal and burning efficiencies, as well as pollutant emissions when evaluating the performance of residential stoves.

IMPLICATIONS

Modern gasifier burners are expected to achieve more efficient burning and consequently lower pollutant emissions, helping alleviate pollution, and greatly improving air quality to benefit human health. Previous laboratory studies, although limited and frequently burning pellets, have reported lower emissions than raw biomass fuel burning in traditional and improved stoves. However, there have been no field studies to evaluate the actual use of these stoves in China. Pollutant emissions usually vary notably among different tests affected by a variety of factors.

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While laboratory tests are used to characterize stove performance systematically under controlled conditions, field measurements can capture emissions from the real burning conditions and provided reliable data for stove evaluation and emission inventory. The present study measured comparable levels of air pollutants emitted from wood combustion in the gasifier stove to those from wood combustion in improved stoves. This indicates the needs for monitoring stove use and pollutant emission measurements, so as to better understand the combustion and emissions of these new burners.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Location of sampling site, pictures of household stoves and fuels, KPT in field, emission factors of organic individuals, calculated emissions factors on a dry basis, and compiled emission data for different fuels in traditional and improved stoves. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/es506343z.

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Notes

The authors declare no competing financial interest.

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