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Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves

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ABSTRACT

We implemented a program in which emission characterization is enabled through collaborations between academic, US and international non-governmental entities that focus on evaluation, dissemination, and in-use testing, of improved cookstoves. This effort resulted in a study of field and laboratory emissions from traditional and improved biofuel cookstoves. We found that field measured particulate emissions of actual cooking average three times those measured during simulated cooking in the laboratory. Emission factors are highly dependent on the care and skill of the operator and the resulting combustion; these do not appear to be accurately reproduced in laboratory settings. The single scattering albedo (SSA) of the emissions was very low in both lab and field measurements, averaging about 0.3 for lab tests and around 0.5 for field tests, indicating that the primary particles are climate warming. Over the course of three summers in Honduras, we measured field emissions from traditional cookstoves, relatively new improved cookstoves, and "broken-in" improved cookstoves. We found that well-designed improved cookstoves can significantly reduce PM and CO emission factors below traditional cookstoves. For improved stoves, the presence of a chimney generally resulted in lower emission factors but left the SSA unaffected. Traditional cookstoves had an average PM emission factor of 8.2 g kg^{-1} – significantly larger than previous studies. Particulate emission factors for improved cookstoves without and with chimneys averaged about 6.6 g kg^{-1} and 4.5 g kg^{-1}, respectively. The elemental carbon (EC) fraction of PM varied significantly between individual tests, but averaged about 25% for each of the categories.

1. Background

Over 2 billion people use biofuels such as wood, dung, and crop residue as their primary means of cooking and heating. Biofuel combustion affects health and environment on a wide variety of scales. Combustion of biofuel is a significant source of particulate matter (PM), producing approximately 20% of global emissions of both organic carbon (OC) and elemental carbon (EC) (Bond et al., 2004). The PM and gaseous emissions cause adverse health effects for the cookstove users and local residents (Ezzati et al., 2002). PM emissions affect Earth's radiation balance both directly and by affecting cloud interactions. Local and regional impacts of aerosol, including visibility reductions and altered weather patterns, can be substantially larger than global impacts (Menon et al., 2002; Ramanathan et al., 2005,2007).

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Despite the importance of cooking emissions for indoor, local, and global air quality, especially in developing countries, they are still not well understood. While numerous studies have measured gaseous and sometimes particulate emissions from standardized or laboratory tests, it is not clear whether these emissions are representative of cookstoves actually in use. Many of these studies use a standardized test instead of simulating real world conditions (Ahuja et al., 1987; Smith et al., 1993; Smith et al., 2000; Zhang et al., 2000; Venkataraman and Rao, 2001; Bhattacharya et al., 2002; Venkataraman et al., 2005).

Although improved cookstoves have been distributed for many years, testing to determine actual impact on users and the environment has only recently begun. Studies in India and Mexico (Chengappa et al., 2007; Masera et al., 2007) show that improved cookstoves reduce the average indoor air concentrations of carbon monoxide and $PM_{2.5}$ generally by 50%. One of the most common performance tests is the water-boiling test (WBT) where a specified quantity of water is brought to boil, and then simmered, while total fuel consumption is recorded. Performance measures include time to boil, energy efficiency, and fuel usage; emissions data can be collected concurrently. The WBT results allow for direct comparison of cookstoves. However, Bailis et al. (2007) found that laboratory measurements do not always translate to similar efficiencies in the field-based kitchen performance test (KPT).

In the project reported here, we sought to answer three questions about cookstove emissions. (1) What are the characteristics of in-use cookstove emissions, and are they similar to those from laboratory studies? In the early phases of the project, we found that they were not (Roden et al., 2006). In-use traditional cookstoves produced more and darker particles than expected from previous laboratory studies. This paper reports a continuation of this inquiry, including a comparison of similar stoves under laboratory and field conditions. (2) Can improved cookstoves ameliorate environmental impacts? Smith et al. (2000) and Zhang et al. (2000) reported that "improved" cookstoves actually produced more particulate emissions per mass of wood than traditional cookstoves. However, that investigation did not examine all types of improved cookstoves. Recent design features such as insulated combustion chambers have not been evaluated. (3) If improved stoves do have lower emissions, do the reductions continue as stoves age?

These questions have implications for interventions designed to address indoor air pollution. They are also relevant to understanding climate and air quality. Total emissions from solid-fuel burning, and optical and chemical properties of particles, are needed as inputs to atmospheric models. Public health issues are now driving implementation of improved cookstoves, and the resulting emission changes need to be understood, both for projecting future emissions and for assessing the success of solutions.

To understand the potential climatic impact, the relevant properties of cookstove emissions must be measured. Aerosols directly affect climate by both scattering and absorbing sunlight. The direct climatic impact is determined by a particle's single scattering albedo (absorption divided by scattering plus absorption). Single scattering albedo (SSA) below 0.85 is generally considered net climate warming (Haywood and Shine, 1995). The SSA is also an indicator of the nature of the particles, and the combustion conditions that created them. Combustion can create both elemental and organic carbon aerosols, but smoldering conditions create primary organic carbon, which appears as white smoke and has SSA between 0.8 and 1. Flaming conditions produce significant amounts of elemental carbon and can have SSA between 0.3 and 0.5. EC creates a large positive climate forcing, which can be amplified when it is mixed with other particles (Bond et al., 2006; Jacobson, 2001).

2. Approach

The University of Illinois participated in a partnership with three non-profit organizations (NPOs) to overcome some critical limitations inherent in the study of traditional biofuel emissions and implementation of improved cookstoves. The interactions between these organizations are diagrammed in Fig. 1. The first limitation is access to sampling locations and obtaining agreement and trust of the cookstove users. In this case, one of the NPOs in the partnership, AHDESA, has a long history of working in Honduras. Because of existing relationships, we were able to field test traditional cookstoves and improved cookstoves at installation and after break-in. The field-testing took place in and around Suyapa, Honduras during the summers of 2004, 2005 and 2006, and was concurrent with a project to disseminate improved cookstoves to the residents.

A second limitation, which is relevant to conducting interventions, is that NPO personnel sometimes do not have a good understanding of testing and measurement issues. This prevents them from assessing the improved stoves. In this partnership, university participants recommended and setup measurement equipment and trained NPO personnel in its operation. Another partner, Aprovecho Research Center, is a U.S. based non-profit that has been developing and disseminating cookstove technology in Central and South America, Asia, and Africa for 25 years. They have greater technical expertise than the average NPO, as well as a need to understand the implications of different stove designs. University participants designed a testing laboratory for Aprovecho that is used for ongoing cookstove research and testing. The laboratory testing enables evaluation of stove performance and emissions while controlling for variables.

Efficiency is an important factor: an improved stove reduces total emissions by the same amount if emission per fuel is decreased by 50%, or if total fuel required for the same task is reduced by 50%. Stove efficiency is usually assessed with the WBT to determine how much of the released heat is transferred into the food. Cooking tasks, such as preparing meals or making tortillas, are not suitable for assessing heat transfer. Our goal in this study was to obtain an estimate of real-world emissions by minimizing interference to normal cooking practices. This constraint

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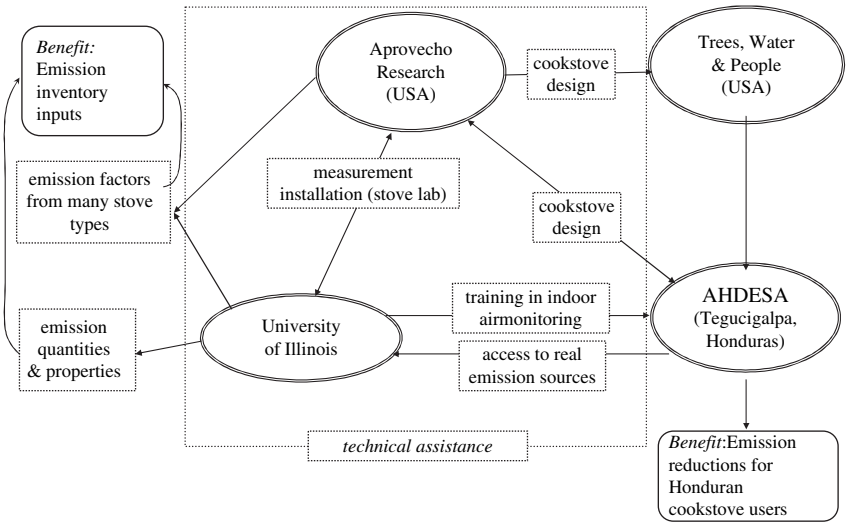

Fig. 1. Diagram of organizations, interactions, and benefits.

made a simultaneous assessment of efficiency impossible for our field tests.

A final limitation is sampling in remote areas where electrical power is unreliable or unavailable. We developed a battery operated sampling system, the ARACHNE (Ambulatory Real-time Analyzer for Climate and Health-related Noxious Emissions), which is described and characterized in Roden et al. (2006). All results in this paper utilized the ARACHNE which measures real-time scattering and absorption by particles with a nephelometer and particle soot absorption photometer (PSAP), respectively, as well as real-time CO and CO_2 gas concentrations. Measurements are recorded every second. These real-time measurements allow us to determine combustion phases that contribute to high PM and EC emissions. Scattering is a proxy for particle emissions, and high absorption relative to scattering is an indicator of elemental carbon (EC) emissions. Particles are also captured on Teflon and quartz filters for analysis.

We made a few slight modifications to the ARACHNE system during the course of the program. In the second year, we added a new CO_2 sensor (Telaire, 6004-S5000) and optimized the flow to minimize its response time. We also added two temperature sensors (National Semiconductor, LM35DM), and a relative humidity sensor (Honeywell, HIH-3610-001). Losses and system performance are discussed in Supplementary material of Roden et al. (2006).

The most important factors in designing the sampling approach were portability and the ability to obtain real-time data. The sampling strategy used here differs from some published source characterization studies, because it does not use a large chamber in which emissions are diluted (Hildemann et al., 1989; Schauer et al., 2001). Such a chamber would require additional power for the necessary blowers, and would severely limit access to sampling locations. For field studies, the equipment was transported in a pickup truck and was often carried to steep, muddy hills. As we will show, emissions from in-use cookstoves differ greatly from those in laboratory tests. We deemed that access to real sources was more important than any uncertainties resulting from a sampling approach that was less formal. A dilution chamber would also limit the ability to obtain real-time data.

Our system relies on the carbon balance method, which is commonly used for sampling open biomass burning (Ward et al., 1996) and biofuel (Bertschi et al., 2003). This approach relies on the ratio between pollutants and a fuel proxy (CO plus CO_2) in the exhaust gas to determine an emission factor (Roden et al., 2006). This method requires a representative sample, so either multiple points in the plume must be sampled equally, or the entire plume must be collected and well mixed before sampling. However, dilution does not affect the emission factors. Samples from stoves without chimneys were taken about 1.5 m above the fire so that initial dilution occurred through natural plume rise and entrainment. Emissions were collected with the Araña probe (Roden et al., 2006), a 24-port collection probe which spans most of the cookstove plume. For chimneyed stoves, one opening of this probe or a diluting probe was placed near the center of the chimney, with the rest of the Araña openings drawing from ambient air, providing an approximate 24:1 dilution. In 2005 laboratory tests, we used the Araña probe inside the stove hood. For 2006 laboratory tests, we used the diluting probe to pull a sample from the hood chimney, which provided a dilution ratio of approximately 3:1 above the naturally diluted hood concentrations and allowed a comparison of repeatability between the two probe types. For field tests of chimney stoves, emissions were well mixed within the chimneys; flow was turbulent ($Re > 4000$), and samples were taken 12–15 diameters downstream of the combustion.

In much source sampling literature, artificial dilution is used to simulate normal atmospheric dilution and to bring emissions to an equilibrium state. Lipsky and Robinson (2005) show that 2.5 s is sufficient for the emissions to

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establish phase equilibrium. Our residence time after dilution varied 3–5 s. The amount of dilution is important in determining the phase partitioning of semi-volatile organics. Lipsky and Robinson (2006) show that too little dilution results in the overestimation of particulate organic emissions. Our dilution ratio of 24:1 is within the typical range of 20:1–200:1. However, the cookstoves operate with an initial dilution of 5–10:1 due to excess air. Our average particle concentrations (2.3 mg m^{-3}) and ΔCO_2 concentrations (760 ppm) correspond to an approximate dilution of at least 100:1 for typical woodstove testing. This dilution ratio, 100:1, is near the maximum total dilution examined by Lipsky and Robinson (2006) for woodstoves. We found no relationship between OC/TC ratio and particle concentration ($R^2 < 0.04$) or between OC/TC ratio and total post-flame dilution (represented by CO_2 concentration, $R^2 < 0.01$). For these measurements, CO plus CO_2 concentrations and PM mass are used to estimate the PM emission factors with the carbon balance method. None of the food cooked during the measurements had high aerosol generation rates, because it consisted primarily of beans, rice, and tortillas.

Emission rates and properties can be impacted by many factors including fuel, lighting, fuel feeding practice, stove type and design, and combustion temperature (Butcher and Sorenon, 1979; Hubble et al., 1981; Muhlbaier, 1981; Butcher and Ellenbecker, 1982; Rau, 1989). Practices varied between laboratory and field in several ways, which are summarized in Table 1. In most of the field emission tests, we tested stoves as used by the residents. We preferred beginning each test with a cold stove, but this was not possible when stoves were in continuous use. If a stove was in use at the beginning of the test, the user extinguished the wood, removed the coals, and then relit the stove after equipment setup (30–45 min of cool down time). In the last year of the study, some tests were repeated with uniform oak wood, which we purchased from a local vendor used by many residents.

Table 2 outlines the type and number of tests performed and the types of stoves tested. The in-use stove tests consisted of thirteen traditional stoves, eight improved ceramic or metal stoves without chimneys, and sixteen improved metal or cement stoves with a plancha (griddle) cooking surface and a chimney. Five of the improved stoves, tested when new in 2005, were direct replacements for traditional stoves tested the prior year. Eight of the improved stoves, which were measured when new (2005), were retested one year later. The small sample size may limit the generalizability of the results; however, our sample, fifty-five in-use tests and fourteen lab tests, is similar in size or larger than previous studies. In the future, automated tests which capture overall emission factors on a large number of stoves should be used to obtain emission factors for sources with such high variability, and to complement the time-intensive, detailed data presented here.

Table 1
Practices affecting emissions

Practice	Field (in-use)	Laboratory
Fuel type	Varied wood: purchased from local vendors, harvested from own or surrounding land, construction debris. Typical size: 5 cm × 5 cm × 70 cm	Dry Douglas fir cut into 1 cm × 2 cm × ~ 30 cm strips
Lighting	Paper, pitch pine kindling, and/or hot coals	Douglas fir kindling and newspaper or lighter fluid if stove unsuited for paper.
Stove initial condition	Cold start, or 30–45 min cool down	Cold start
Cooking practice	Operators' meal	Standardized water-boiling test

Table 2
Description of the types of stove tests, the setting and year tested

	Stove type	2004	2005	2006	'06 (our oak)
Field	Traditional	11	2	0	
	New improved without chimney		6		
	New improved with chimney		13		
	Broken in improved without chimney			5	3
	Broken in improved with chimney	1		7	7
	Field retest of 2004 homes		5		
	Field retest of 2005 homes			8	
Lab	Traditional		1	1	
	Improved		3	2	
	Gasifier		1	2	
	Fan		0	4	

Laboratory results were collected during two separate workshops at Aprovecho in 2005 and 2006 utilizing the ARACHNE. At these workshops, participants bring or build improved cookstoves, which are usually simpler single-pot stoves without chimneys. These stoves are tested for emissions while performing the water-boiling test. The stoves tested include a traditional three-stone fire, traditional ceramic or mud one pot stoves, improved one pot stoves, gasifiers, and fan stoves; it was not possible to test exactly the same group of stoves in the field and in the laboratory. Pictures of some of the field and lab stoves are included in Supplementary material. The first set of laboratory tests did not include the PSAP, or quartz filters, so no absorption or EC data is available for those tests.

System reproducibility and the impact of different probes were examined with tests of three identically designed, improved cookstoves measured at Aprovecho under similar operating conditions. For these tests, two with the Araña, and one with the diluting probe, the coefficient of variation is 4% and 13% for PM EF and CO EF, respectively. Users cooked different foods throughout the day and used varied types and sizes of wood, so we could not use repeated tests in Honduras as measures of reproducibility.

The terms "traditional" and "improved" are ubiquitous in cookstove descriptions; however, all "improved" stoves are not equal. "Improved" in this context (both laboratory and field) indicates a combustion chamber constructed of a lightweight refractory material and surrounded by insulation (Still et al., 2000). Most of these improved stoves

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have been designed to optimize airflow through the stove and heat transfer to the pot. Only a few of the "improved" stoves reported by Smith et al. (2000) and Zhang et al. (2000) had insulated combustion chambers, and the design for optimal airflow through the combustion chamber was not detailed. The importance of a lightweight, insulated combustion chamber was identified only after many early stove programs were underway.

3. Results and discussion

Results from field and laboratory tests, categorized by stove type (traditional, improved without chimney, improved with chimney) and testing conditions (in-use field and standardized lab), are summarized in Fig. 2. These summaries do not include the field tests where our provided wood was used. A table providing results of individual emission tests is provided in Supplementary data. This table lists the stove type, the probe used, emission factors, the elemental carbon (EC) to total carbon (TC) ratio, and the average single scattering albedo for each test.

Fig. 2A illustrates that PM emission factors (EFs) are largest for traditional in-use cookstoves. The average PM emission factors for improved cookstoves with chimneys (4.5 g kg^{-})_was almost 50% lower than that of traditional cookstoves (8.2 g kg^{-}); this difference is statistically significant ($p = 0.005$). Improved cookstoves without chimneys also had somewhat lower PM emission factors than traditional cookstoves, but this difference was not statistically significant ($p = 0.168$).

The average of all field PM EFs (6.1 g kg^{-}) is four times larger than that from laboratory tests (1.5 g kg^{-1}). A better comparison is between similar sets of stoves. Improved non-chimney stoves (field) and single-pot traditional and improved stoves (laboratory) have means of 6.6 g kg^{-1} and 1.8 g kg^{-1}, respectively, and the difference is statistically significant ($p < 0.001$).

The mean PM emission factor for all in-use cookstoves is two to three times larger than those measured in previous laboratory studies of traditional and improved cookstoves (Venkataraman and Rao, 2001; Zhang et al., 2000). These studies, which were performed either in a laboratory setting or a simulated kitchen, are shown in Fig. 2A. The EFs are similar to our laboratory data. Li et al. (2007) reported an average PM EF for Chinese improved cookstoves of 3.3 g kg^{-1} of wood performing standardized water-boiling tests. This finding is important; it implies that laboratory tests do not fully capture some of the important factors that lead to particulate emissions. The much lower variability for laboratory PM emission factors compared to field measurements will be examined below.The average of the two traditional cooking fires sampled in the lab (2.1 g kg^{-1})

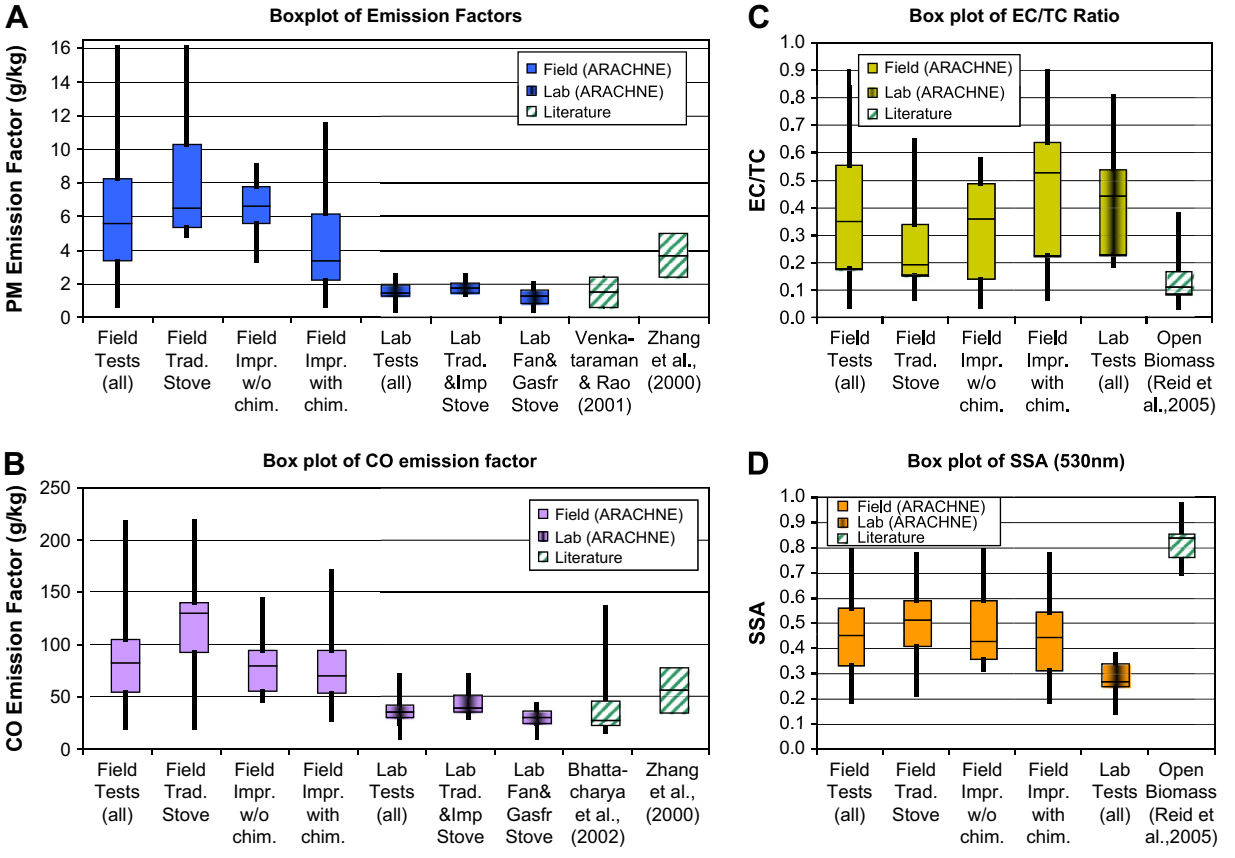

Fig. 2. Box plots of field, lab, and literature results for (A) PM emission factors, (B) CO emission factors, (C) EC/TC ratio (D) SSA. Venkataraman and Rao (2001) and Zhang et al., (2000) results are shown as average ± one standard deviation instead of box plot.

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was slightly higher than the overall laboratory average; however, further comparisons with field traditional stoves are difficult due to the small sample size.

Carbon monoxide emission factors, shown in Fig. 2B, generally follow the same trends as the PM emission factors. Traditional cookstoves have the highest average CO EF, 118 g kg^{-1}. The average for all improved cookstoves, 76 g kg^{-1}, is significantly lower, and the difference is statistically significant ($p = 0.033$). These field-based values are larger than previous laboratory studies by Bhattacharya et al. (2002) and Zhang et al. (2000), which average 42 and 56 g kg^{-1}, respectively, for combined traditional and improved wood-burning cookstoves. They are also larger than our laboratory-based CO emission factors: 44 g kg^{-1} for traditional and improved cookstoves, and 29 g kg^{-1} for fan and gasifier cookstoves. However, these studies, again, used a standardized WBT. Our improved in-use cookstove average is similar to the 63.6 g kg^{-1} average found by Li et al. (2007) who used a standardized WBT.

Figs. 2C and 1D show the elemental carbon to total carbon ratio (EC/TC) and SSA for different stove types and testing conditions. All EC/TC ratios are significantly larger than those found for open biomass burning which averages around 0.1 (Reid et al., 2005). The cookstove EC/TC ratio is closer to the ratio of diesels, which typically ranges from 0.52 to 0.78 (Gillies and Gertler, 2000). The lab tests generally have a lower SSA than the field tests. These measurements illustrate that cookstove emissions are not similar to those from open biomass burning. The EC fraction in cookstove emissions is significantly larger, as also shown by Venkataraman et al. (2005), and the SSA averages 0.4–0.5 instead of 0.8 for open biomass burning.

EC/TC measured in the field varied widely with traditional stoves having the lowest average (0.24), and improved stoves with chimneys having the highest average (0.52). The EC/TC ratio of the lab tests was also fairly high with an average value of 0.45. The average EC EF was largest for improved stoves without chimneys. The average EC EF was similar for traditional and improved stoves with chimneys, but the improved stoves with chimneys showed the greatest variability among all stove categories. The OC and TC emission factors followed the same patterns as the PM EF between the stove categories. The median single scattering albedo is lower for improved stoves than for traditional stoves.

3.1. Specific household comparisons

3.1.1. Traditional versus improved stoves

We tested five homes with traditional cookstoves, and then retested them once an improved cookstove with a chimney was installed. Fig. 3 illustrates this comparison for PM emission factor, SSA, EC fraction, and CO emission factor. The initials under each bar graph (J, ES, EL, Eco_H) indicate different stove types, which are described in the supplemental data. PM emission factors were reduced by the improved cookstove in all cases, with the average reduction over 70%, and a paired t-test yielding a p-value of 0.021. The average EC/TC ratio did not change. The CO emission factor was generally 20 to 50% lower for improved

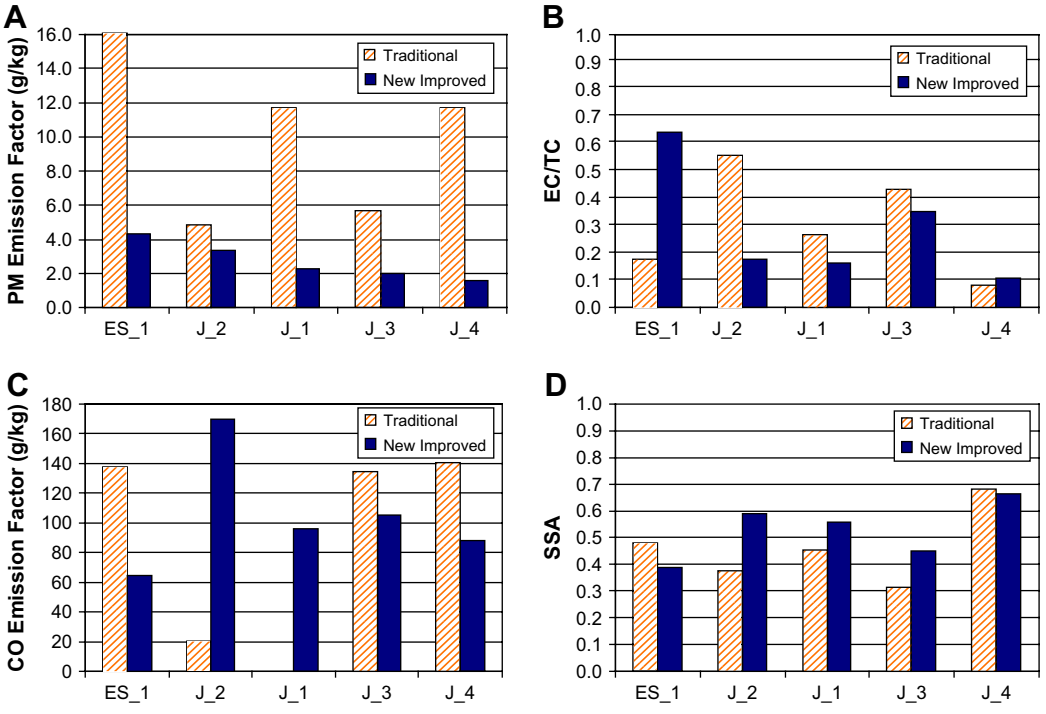

Fig. 3. Comparison of traditional stoves versus improved stoves with chimneys in the same households. ES and J indicate the type of improved stove that replaced the traditional stove. Test J_1 did not have CO data due to equipment problems.

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stoves; however, for one of the improved stoves (J_2), the CO EF was unexplainably much larger. Paired *t*-tests indicate no statistical difference ($\alpha = 0.05$) between the SSA, EC/TC and CO for these five pairs of cookstoves.

3.1.2. Effect of one-year break-in and wood type

In 2006, we tested how a one-year break-in period affected stove emissions. Emissions might decrease as the user gained familiarity with stove operation, or they might increase as the stove degraded. During this final year, we also explored reasons for the high emission variability observed in previous years, including the effect of wood type. In some cases, we performed two cooking tests, one with the homeowner's wood, and one with oak purchased from a local wood vendor. We chose oak because preliminary results indicated that its emissions were lower than other wood such as pine. This purchased oak had an average moisture content of $31 \pm 8\%$ on a dry wood basis, and we cut it to fairly uniform pieces of approximately 2 cm × 2 cm × 70 cm. The wood normally used had a larger cross-section of about 5 cm × 5 cm, and had average moisture of $18 \pm 6\%$ (on a dry basis).

PM emission factors for the same stove under varying conditions (new, one-year break-in, one-year break-in with provided wood) are shown in Fig. 4. The data are separated into stoves with and without chimneys. Averages and standard deviations are also shown for PM EF, SSA, EC/TC ratio, and CO emission factor. PM emission factors generally increased after the stove had been used for one year, with an average increase of 50%. This difference is not statistically significant ($p = 0.115$ from a paired *t*-test). When the same stoves were tested with the oak that we supplied, the PM emission factors had a significant average reduction of 50% ($p = 0.019$). The variability in PM EF for chimney stoves was low when identical fuel was used suggesting that much of the variability is due to wood type and size. One stove without a chimney (EL_1) had high emission factors under all three conditions. This was primarily due to poor lighting procedure, which caused large smoldering during ignition.

Previous studies have contradictory results regarding the impact of wood type on fireplace emission factors. McDonald et al. (2000) found PM emission factors from fireplaces were approximately the same for softwood (ponderosa and pinion pine) and oak. However, Fine et al. (2004) showed that white oak produced particulate emission factors twice that of Douglas fir and pine for both fireplaces and woodstoves. Schauer et al., (2001) found the opposite that fireplaces burning oak produce half the particulate emission factor when compared to pine. Studies

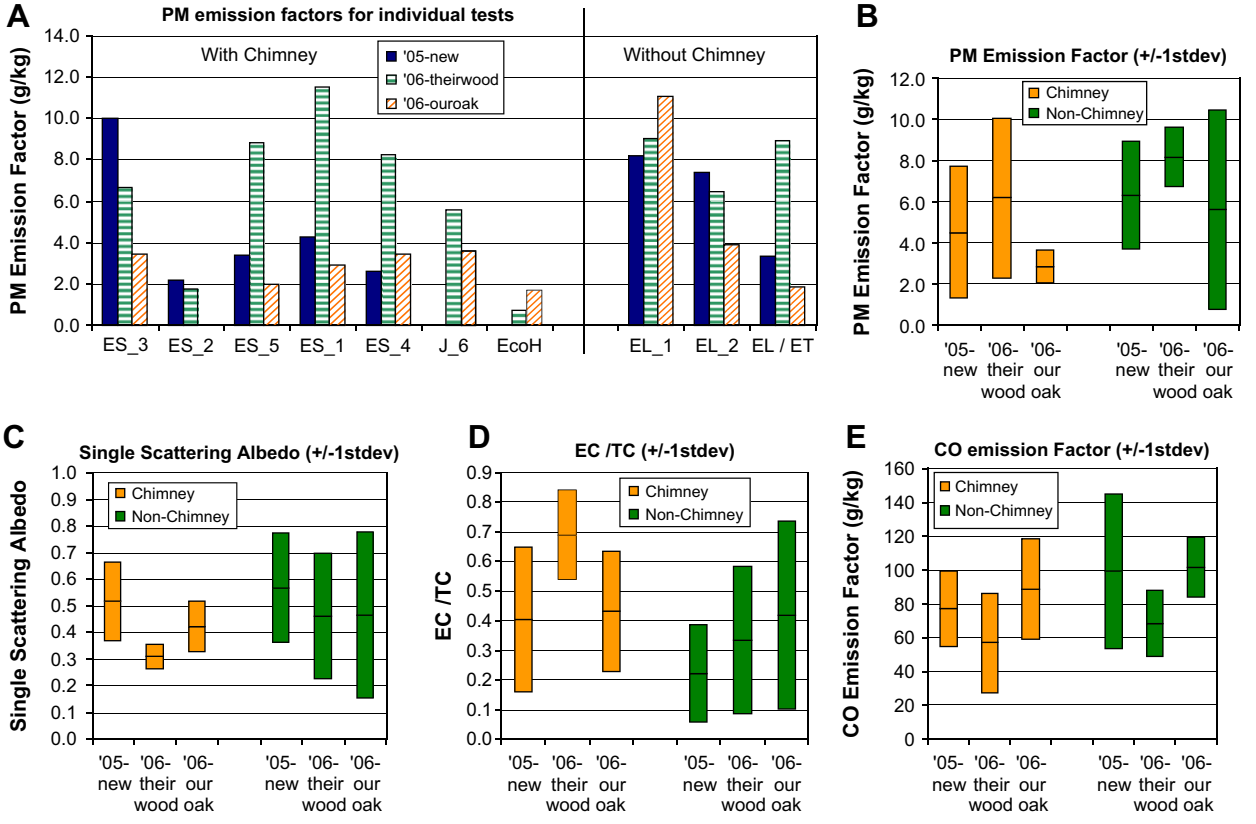

Fig. 4. Comparisons of stoves that were measured under three different conditions (new, one-year old, and one-year old with provided oak). (A) PM emission factors of individual stoves tested under different condition. ES, J and Eco_H are different improved stoves with chimneys, and EL and ET are improved stoves without chimneys. (B–E) The average ± one standard deviation for improved stoves with and without chimneys under different conditions (B) PM EF, (C) SSA, (D) EC fraction and (E) CO emission factor. In (A) Stove ES_2 was not retested with our oak, and two stoves (J_6 and EcoH) were not tested when new, but only after the one-year break-in period.

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on the influence of fuel size are also inconclusive. For fireplaces, a smaller log size (split versus halved) resulted in a 40% and 70% reduction in PM emission factor for pinion pine and oak, respectively, (Muhlbaier, 1981). The larger log sizes resulted in a decreased burn rate and decreased temperature. Barnett and Shea (1981) showed that for the same mass wood loading, smaller diameter logs (“1” diameter versus “8” diameter) resulted in a 350% increase in PM emissions.

Results from North American wood heating stoves and fireplaces are expected to be different than those of cookstoves. Heating stoves are often loaded less frequently. Cookstoves have much smaller combustion chambers, and the heat is directed toward the cooking pot, so operators must regularly add fuel in the front. The result is a more continuous feed. For cookstoves, smaller diameter fuels allow the user greater control of the fire, ranging from simmer to high power. Smaller diameter fuels are easier to keep lit, but also require more attention due to regular fuel additions. The ease of lighting and keeping the fuel lit reduces the amount of smoldering aerosols which can dominate emissions of poorly operated cookstoves. However, more work is involved in cutting the fuel to achieve the smaller diameter, so users generally prefer to use the wood as received.

The mean CO emission factor decreased by 24% after the cookstoves were used for at least one year. However, this decrease is not statistically significant at $\alpha = 0.05$. In tests with the provided oak, the average CO emission factor increased 50% when compared with the same tests using the residents’ drier wood. A paired t-test indicates that this difference is statistically significant ($p = 0.002$). The increased CO emission factor for wet fuel is consistent with other studies. Bhattacharya et al. (2002) showed that increasing moisture from 10 to 25% (on dry basis) resulted in a 40–90% increase in CO emission factor for paired tests on traditional and improved cookstoves.

The EC/TC ratio was the largest, and the SSA was the lowest for broken-in, improved cookstoves with chimneys that were tested with the owners’ own wood. This fuel often consisted of pine construction debris and pine logs, some with high resin content. Similarly, McDonald et al. (2000) found that EC fraction for softwood (ponderosa and pinion pine) emissions were twice that of oak, although their EC fractions were significantly lower than ours. Improved cookstoves without chimneys displayed much variability in the SSA and EC/TC ratio, but did not exhibit any clear patterns or trends.

3.2. Emissions resulting from lighting and fuel addition

Cookstove ignition and fuel addition both emitted large quantities of particulate matter, observed as spikes in the scattering and absorption signals, which are accompanied by smaller increases in the CO_2 and CO signals. These perturbations generally lasted between three and five minutes. Butcher and Sorenon (1979) also reported that heating stoves produced the majority of emissions during startup. To examine the impact of lighting on emissions, we averaged the data produced during the first five minutes of each test, and compared this “lighting average” with the average during the entire test to determine the “lighting emission ratio”. Lighting emission ratios for CO, CO_2, scattering (a proxy for particulate matter) and absorption (a proxy for elemental carbon) are presented in Table 3. Emissions caused by fuel addition may also occur later in the test. Peaks in the real-time data were invariably associated with fuel addition, recorded either by direct observation or by an automated digital camera. However, because many small fuel addition or adjustment events occurred during each test, we were not able to quantitatively apportion emissions to these events.

For in-use cookstoves, the first five minutes of a test produces between two and three times the mean scattering and mean absorption, while the CO_2 emissions (representative of fuel burning rate) are only half that of the overall average emissions. In laboratory tests, the average lighting emission ratio for both scattering and absorption ($\sim$200%) is comparable to the field averages. However, the lab lighting emission ratios for CO and CO_2 are significantly larger than those for the field tests. This is likely due to the use of dry, smaller, uniformly sized fuel which takes less time to ignite and establish a stable fire. The field tests using our provided oak produced the highest average lighting emission ratio for both scattering and absorption ($\sim$300%); this is probably due to the higher moisture content of the purchased oak, coupled with lower emissions during the average use. For individual tests, there were no correlation between the scattering or absorption lighting emission ratio and the overall PM emission factor of the test.

To further explore the contribution of single events to total emissions, we examined the distribution of real-time measurements (10 s averaging time) for scattering, absorption, CO_2, and CO. Scattering, absorption, and CO appear more lognormal than normal, with skewness ranging from about 1 to 4 for scattering and absorption and from 1 to 5 for CO. However, statistical tests indicate that these distributions are not lognormal, either. Table 4 summarizes the contribution of high-emitting events for each stove type, by averaging the fraction of the time during which the signal was above the mean. If the signal had a normal distribution, half the measurements would fall below the mean, while a smaller fraction above the mean indicates that high events contribute greatly. For both lab and field data, approximately 70% of the real-time scattering and absorption measurements fall below the mean. For CO_2, about half of the measurements are above the average, suggesting CO_2 has a more normal distribution. For individual tests, there was no correlation between

Table 3
Lighting emission ratio (lighting average/test average) for CO, CO_2, scattering, and absorption

	CO (%)	CO_2 (%)	Scattering (%)	Absorption (%)
Lab (all stoves)	109	116	199	234[a]
Improved stove (our oak)	54	33	322	305
Improved stove (user's wood)	61	56	184	216
Traditional stove	82	68	204	184

[a] Lab absorption data does not include 2005 lab test.

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Table 4
Fraction of real-time data points falling above the signal mean for CO, CO_2, scattering (530 nm) and absorption (530 nm)

	Fraction of data above signal mean			
	CO	CO_2	Scattering	Absorption
Lab (all stoves)	0.40	0.47	0.31	0.30[a]
Improved stove (our oak)	0.38	0.43	0.24	0.22
Improved stove (user's wood)	0.39	0.47	0.28	0.28
Traditional stove	0.46	0.48	0.32	0.27
Minimum for all tests	0.23	0.25	0.09	0.14
Maximum for all tests	0.57	0.61	0.48	0.38

[a] Lab absorption does not include tests performed in 2005 since no absorption was measured.

the percent of scattering, absorption, CO, or CO_2 above the average and the emission factor. Thus, predictive relationships are not possible, but this summary points to the general importance of lighting and fuel addition in producing particulate emissions.

Fig. 5 demonstrates the asymmetry of the scattering signal from a single test. In this figure, 10 s data slices are sorted (largest to smallest) and then plotted against cumulative test time. In this typical case, scattering is greater than the mean 28% of the time, and only 10% of the emissions, on a time basis, results in half of the cumulative scattering. In the most extreme case, half of the cumulative scattering occurs during 5% of the time, with only 9% of the scattering above the mean. This illustrates how a few short duration events can dominate the overall scattering emission leading to high PM emission factors.

3.3. Field versus standardized laboratory testing

One major finding is the difference between in-use field emissions and laboratory measurements based on a standardized test. Two major differences between these testing conditions are the fuel variability and the fire tending. All lab-based tests used identical fuel (Douglas fir cut into 1 cm × 2 cm × ~ 30 cm strips) whose small cross-section allowed for precise metering. During laboratory tests, the cooking fire was constantly tended, and fuel was added more frequently, but slowly and gently. During in-use tests, the primary focus is not the cooking fire; cooks must divide their attention between food preparation, children, other household tasks, and fire tending. Two representative real-time plots of absorption, scattering, SSA, CO and CO_2 versus time are given in Fig. 6. These tests are of similar improved cookstoves with no chimney. The bottom (in-use) graphs show rapidly fluctuating emissions. SSA fluctuates between periods of dark smoke (SSA ~ 0.3) from flaming combustion and periods of smoldering combustion (SSA ~ 1). The laboratory tests (top) show much less variability. The fairly stable SSA (~0.3) suggests smoldering rarely occurred. The CO and CO_2 signals are also much less variable, probably because of the consistent wood feed rate.

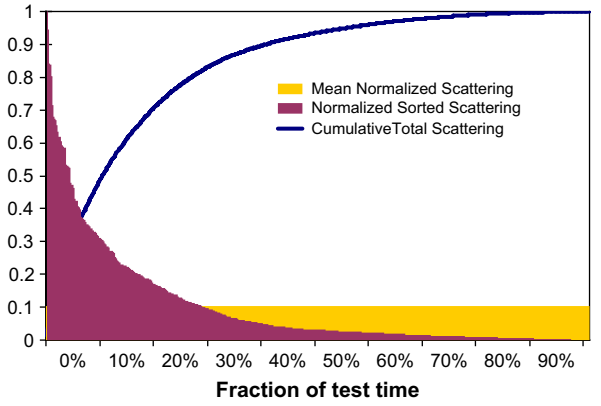

Fig. 5. Sorted scattering, and cumulative total scattering as a function of total test time from a typical test (test 323 – improved stove without chimney, PM EF = 4.7 g kg^{-1}). "Sorted normalized scattering" and "mean normalized scattering" are normalized to the single largest scattering measurement.

We utilized statistical analyses and signal processing techniques on the real-time data to quantify the difference between the laboratory and field tests. Several of these analyses found statistically significant differences between field data versus the laboratory data. However, we were generally thwarted in the effort to predict PM emission factor based on these analyses; a brief description of these attempts is given below.

We first examined the standard deviation of the real-time SSA, and the coefficient of variation (standard deviation divided by mean) for the real-time scattering, absorption, CO_2, and CO concentration measured during each test. The standard deviation of SSA is used instead of coefficient of variation since SSA is a ratio of two signals. Table 5 gives the laboratory and in-use means of the coefficient of variations, as well as the p-value for t-test comparing the difference between lab and field data sets. The results show a statistically significant difference ($\alpha = 0.05$) for all values except scattering, whose p-value is only slightly above 0.05. These results reaffirm that the standardized tests did not reproduce the variability observed in the field tests. However, for individual tests, neither the coefficient of variation for CO, CO_2, scattering, nor absorption are correlated with the PM emission factor.

We used a Fast Fourier Transform on the real-time CO, CO_2, scattering, absorption, and SSA to determine the energy spectral density for each signal. For the CO and CO_2 signals, the lab tests contained a smaller fraction of spectral energy at high frequencies (less than 5 min) when compared to the field tests; however the results were not statistically significant. There was essentially no difference between the lab and field mean energy for absorption and scattering. We also found no significant relationship between the fraction of spectral energy in different frequency ranges and either PM emission factor or EC/TC ratio.

Lastly, we determined the number of transitions between flaming and smoldering using the SSA, defining flaming as SSA below 0.35 and smoldering as SSA above 0.80, and counting only data with significant emissions (extinction greater than 500 Mm^{-1}). On average, the lab tests had 2.5 transitions per hour, and the field tests averaged 6 transitions per hour. This measure yielded the

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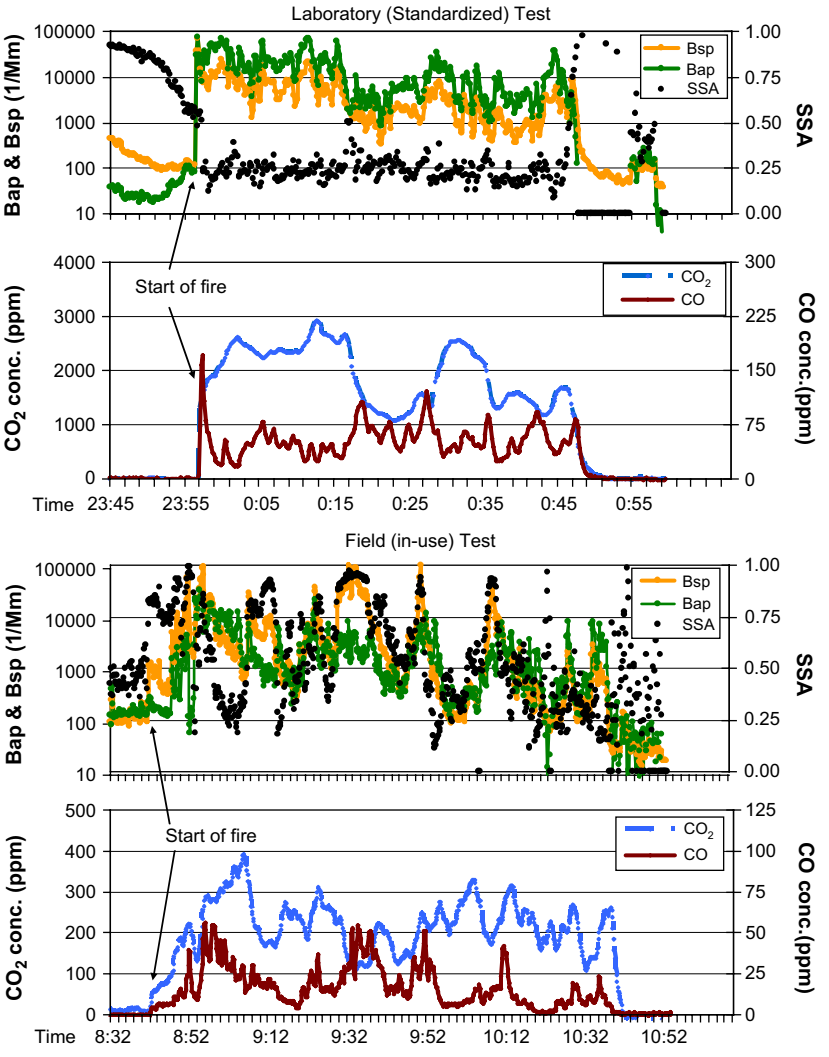

Fig. 6. Real-time scattering (Bsp) in yellow, absorption (Bap) in dark green, SSA as black points, and in separate plots CO_2 in dashed light blue, and CO in dark solid red. The bottom two plots are from field test 308 (improved no chimney cookstove) PM EF = 9.0 g kg^{-1} CO EF = 80 g kg^{-1}. The top two plots are from lab test 410 (improved no chimney cookstove) PM EF = 1.4 g kg^{-1} CO EF = 30 g kg^{-1}.

greatest distinction between in-use versus laboratory results, partly because many of the lab tests did not have a single transition, operating exclusively in the flaming mode throughout the test. Transitions per hour did correlate weakly with the PM emission factor for the laboratory data and the 2006 field data with provided oak, with R^2 of 0.32 and 0.48, respectively. However, there was no correlation for the other field data, suggesting that other factors are more critical in determining emission factors.

3.4. CO EF versus PM EF

Carbon monoxide emissions are sometimes used as a proxy for PM emissions, since both pollutants are formed from incomplete combustion. Fig. 7 plots PM emission factor against CO emission factor to examine this assumption, which proves fairly valid for the laboratory tests ($R^2 = 0.58$). However, there is essentially no correlation for in-use stoves where fuel type and combustion conditions varied widely from user to user ($R^2 = 0.09$). Some tests produced large particulate emission factors with low CO emission factors and vice versa, suggesting that CO is not a valid proxy for PM for field measurements of cookstoves.

Table 5
Coefficient of variation for real-time scattering, absorption, CO_2, and CO, and SSA standard deviation

	Average coefficient of variation				Standard deviation
	CO	CO_2	Scattering	Absorption	SSA
Lab (all stoves)	0.65	0.42	1.38	1.30[a]	0.12[a]
Field (all stoves)	0.87	0.60	1.68	1.77	0.19
P-*t*-test (unequal variances)	0.008	0.002	0.054	0.006	0.001

[a] These values only include the 2006 lab results since absorption was not measured during 2005 lab tests.

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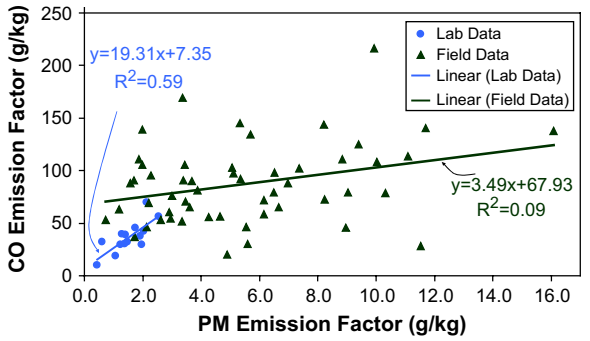

Fig. 7. PM emission factor regressed against CO emission factor, separated into laboratory and field data.

CO might be a valid proxy for PM when non-combustion factors, such as fuel switching and chimneys, are dominating emissions (Naeher et al., 2001). However, when variables such as wood type and stove design contribute strongly to emissions, CO should not be used to represent PM.

4. Conclusion

We describe a new approach for expanding the database of climate-relevant emission characteristics: "piggybacking" on existing projects and collaborating with non-profit organizations. Without the many organizations involved in this partnership, the results presented here would not have been possible.

Well-designed improved cookstoves can significantly reduce PM and CO emission factors below traditional cookstoves. Variables such as lighting procedure, fuel addition, and wood size and type are also significant in determining emission factors. The average of all field measured PM emission factors is over three times as large as laboratory measured PM EF using the same sampling methodology. In laboratory tests, the use of consistent fuel and careful stove operation results in much less emission variability.

Lab tests serve a useful purpose in comparing relative performance of different stoves under similar conditions. However, emissions from laboratory testing should not be considered representative of real-world emissions, at least until the important variables are identified and represented. Therefore, we advocate caution in using laboratory tests to estimate atmospheric emissions. This has ramifications for other standardized tests used in characterizing emissions from wood-burning fireplaces and heating stoves. To promote a better understanding of real-world emissions, field-testing needs to identify the critical conditions and variables governing emissions, and laboratory testing needs to be designed to emulate these conditions.

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Appendix. Supplementary information

Supplementary data associated with this article can be found in the online version at doi: 10.1016/j.atmosenv.2008.05.041.

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